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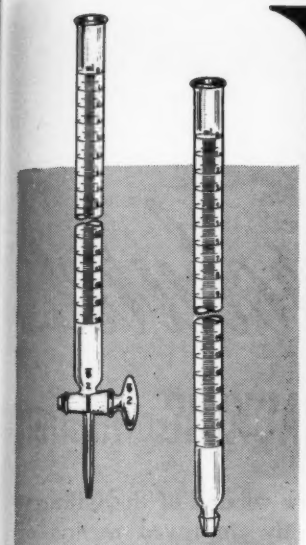

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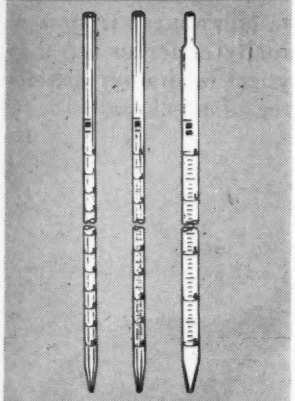

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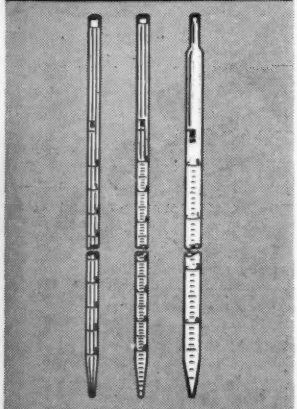

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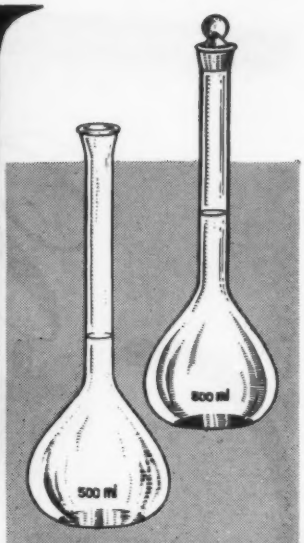

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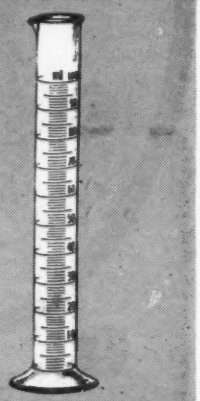

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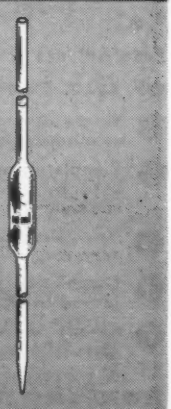

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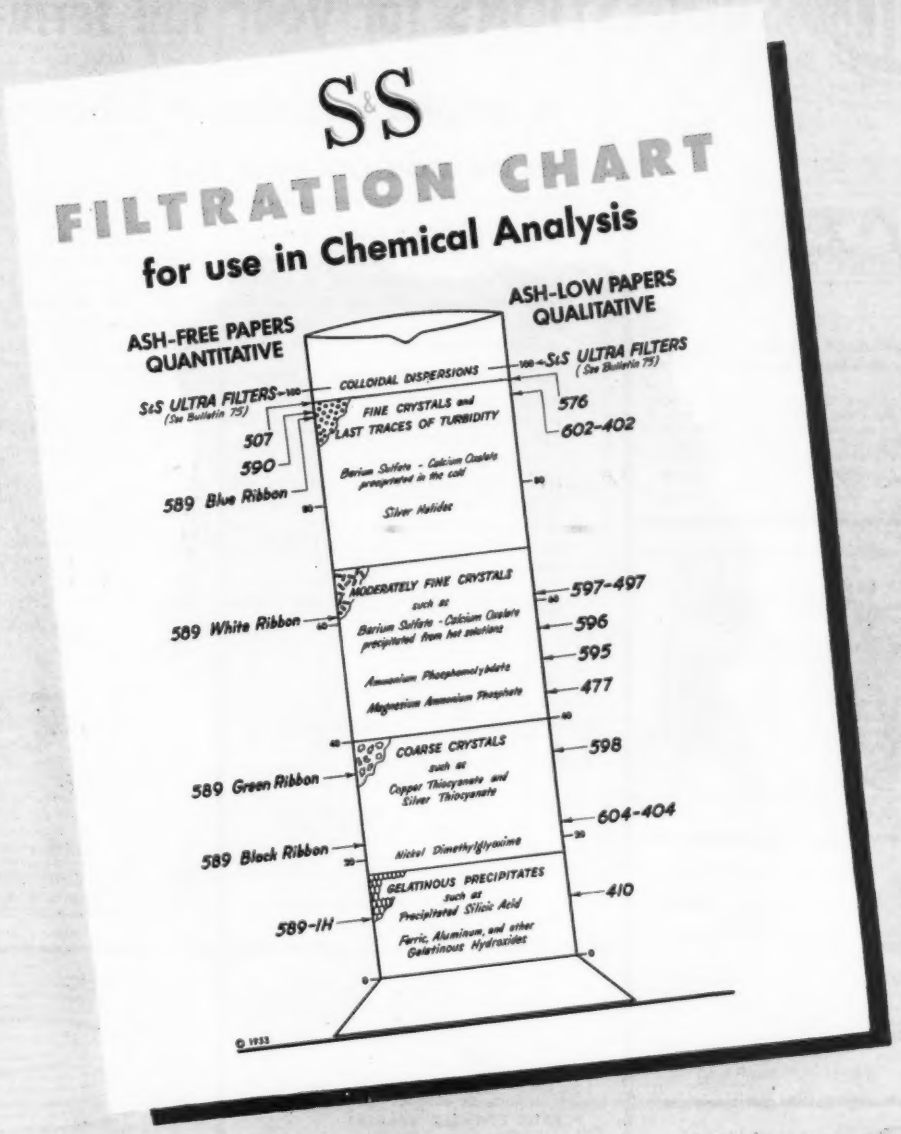

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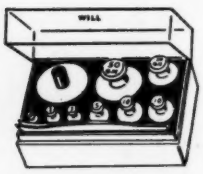

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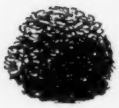

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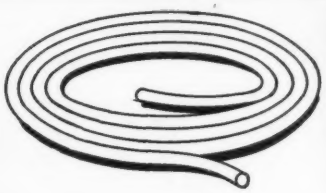

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Odorless, tasteless, nontoxic and chemically inert—even in presence of highly corrosive reagents such as NaOH, hydrofluoric and sulfuric acids. Withstands flexing almost indefinitely. Easily attached to, and removed from, glass tubing. Wall thickness, 1/16".

23525T

I.D. inches	1/8	3/16	1/4	3/8	1/2
Per foot	$.21	$.28	$.32	$.38	$.46
Per 100 ft.	$17.	$22.	$25.	$32.	$40.

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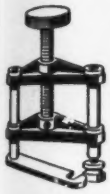

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For large or heavy wall tubing

Recommended for use with large diameter or heavy nitrometer or pressure tubing. Size of opening, 35mm. wide, 30mm. high. Made of brass throughout, nickel-plated.

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$17.76 doz.

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For quick, easy adjustment

(Varigrip) Stainless steel. Flexible band with worm-drive thumbscrew adjustment, easily holds cylinders from 2¾" to 6½" O.D.

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For handling glass tubing safely

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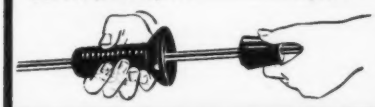

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per 50 ft. roll... 47.25

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(Static Master) Cleans balances, microscopes, film and optical apparatus. Removes dust by destroying static surface attraction. Utilizes harmless radio-active Polonium. Guaranteed for 1 year. Width 1".

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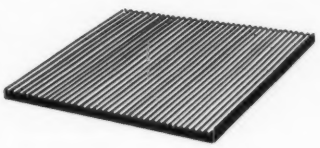

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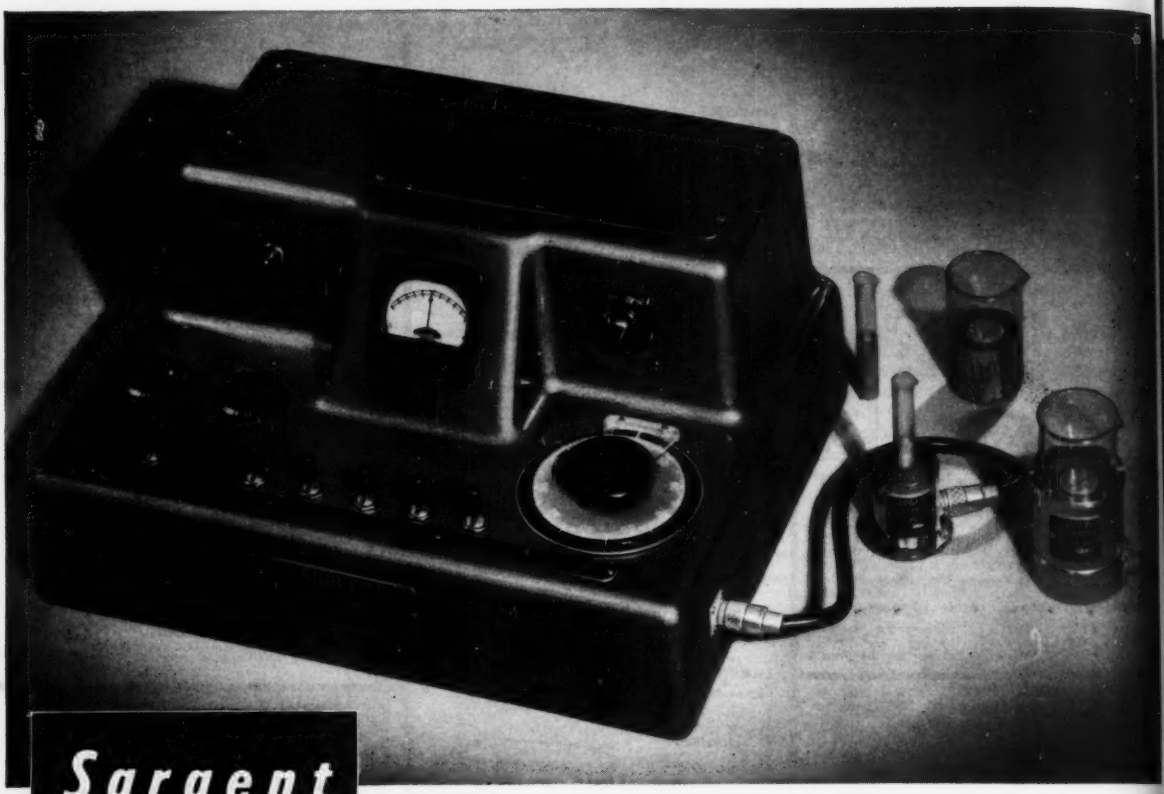

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A very sensitive, stable measuring instrument for chemical analytical procedures, based on the electrical properties of the substances to be identified, differentiated or determined. Opens a new broad field for the development of analytical methods and control procedures.

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In its measurement of characteristics, it recognizes resistivity, electrical moments of the molecule and all factors contributing to the dielectric constant.

Unlike prior electrometric instruments for chemical purposes, the Oscillometer operates with the samples isolated in glass cells, having no structural connection of any kind with electrical circuits, and so precludes any possibility of electrolytic alteration of sample composition and of contamination by diffusion effects or metallic solubilities.

In terms of dielectric constant as applied to the measurement of non-aqueous substances, the sensitivity and reproducibility of the Sargent Chemical Oscillometer averages about 1/300 (one three-hundredth) of a dielectric constant unit, when using full range cells. Stated otherwise, the range of dielectric measurement from air to water corresponds with a scale range in the oscillometer of roughly twenty-four thousand units, reproduceable within approximately one such scale unit, insofar as instrumental characteristics are concerned.

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The detection of water in non-aqueous media is extremely sharp and indicates direct procedures in many instances.

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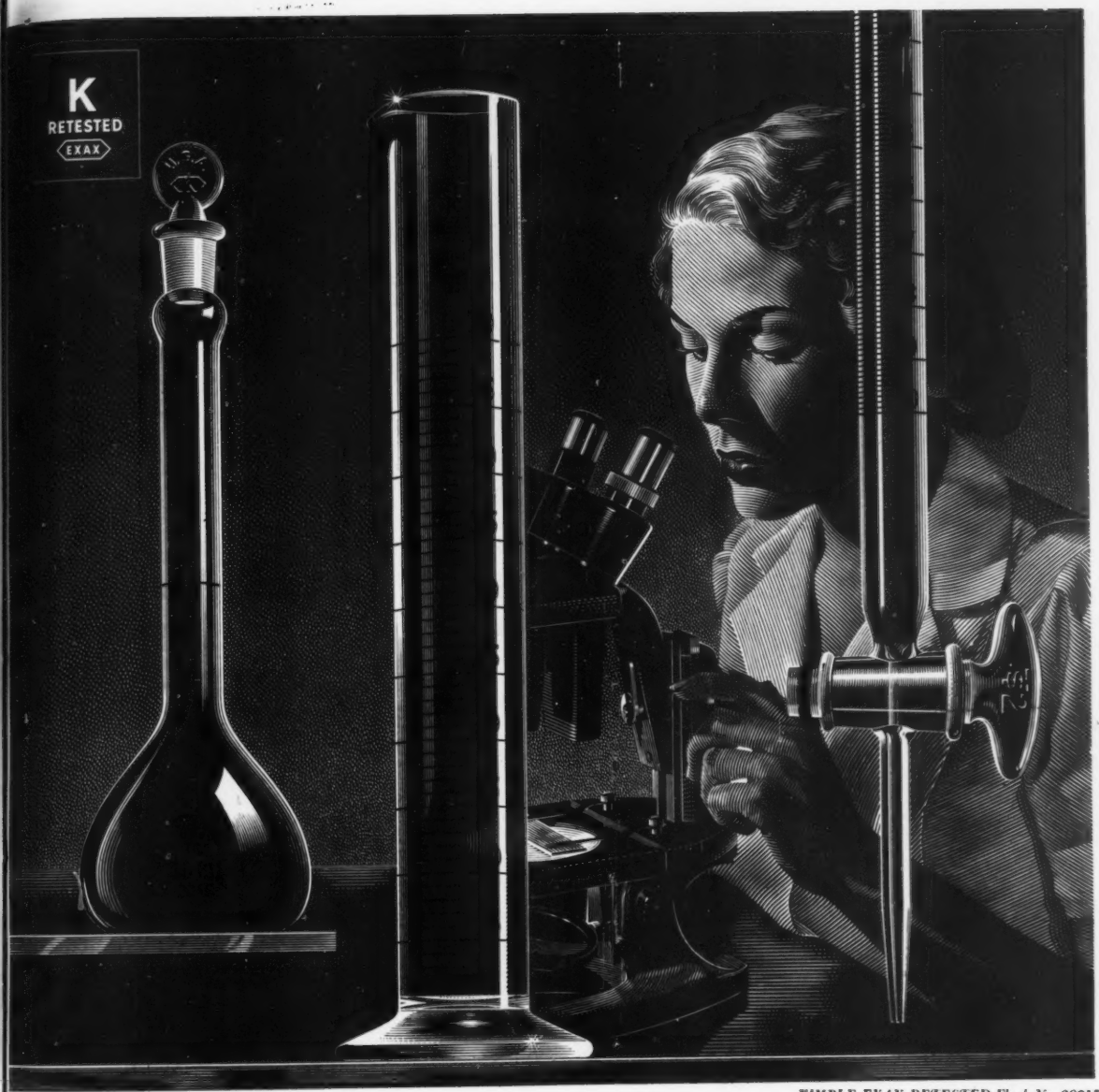

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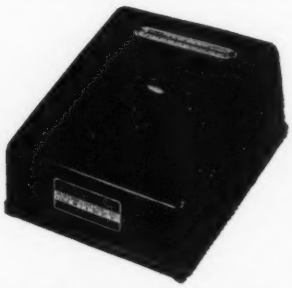

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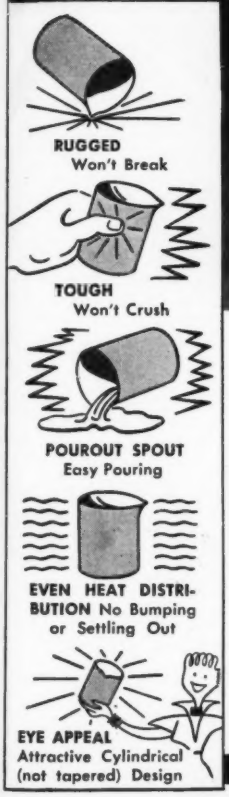
250 ml	126 gm	2-1/2	3-5/8	2.10	1.90
600 ml	242 gm	3-5/16	4-3/4	2.75	2.50
1200 ml	325 gm	4-1/16	5-5/8	3.40	3.05
2000 ml	446 gm	5-1/16	6-1/4	4.20	3.80
3000 ml	586 gm	5-7/8	6-7/8	4.75	4.25
4000 ml	691 gm	6-1/2	7-7/8	5.45	4.90

(GRIFFIN LOW FORM)



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No. CE 192-88
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150 ml	52 gm	$1.90	$20.52
250 ml	72 gm	2.10	22.68
500 ml	105 gm	3.10	33.48
750 ml	135 gm	4.10	44.28
1000 ml	169 gm	4.50	48.60

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When Versene is added to a mixture of various metallic ions one metal will be complexed before another. After the first is exhausted the second will be chelated and the process will continue until either metallic ions or Versene are consumed. There are seven Versenes and all of them have selectivity of complexing power. Some, however, are much more aggressive toward certain metals under special conditions than others.

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When you want to conquer foreign metallic ions in solution — investigate *all* of the Versenes. Made only by the Bersworth Chemical Company, under processes originated, developed and patented by F. C. Bersworth, they are unduplicated in quality and uniformity of complexing power. Whatever your problem in chelation may be, our quarter-century of pioneering the polyamines can help you. Technical Bulletin #2, samples and competent chemical counsel available on request and without obligation.

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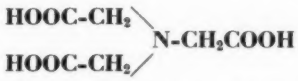
Versene® CHELANTS for HEAVY METALS

Under formulas originated and developed by F. C. Bersworth, the Bersworth Chemical Company manufactures a series of chelating agents (chelants) whose complexing power is effective in varying degrees for heavy metal ions. Versene-NTA is now available in experimental quantities for experimental purposes. Versene Fe-3 Specific and Versene-EDG are available in commercial quantities. We are glad to define them and their properties as shown:

VERSENE-NTA

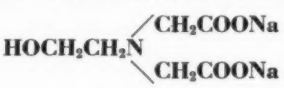
This is nitrilotriaceticacid. Its chemical structure is represented as follows:



Versene-NTA salts effectively chelate heavy metals and alkaline earth ions. But its complexing power compared to Versene itself is considerably less. Although it complexes trivalent iron in acid solutions, extensive hydrolysis of the metal chelate greatly reduces its effectiveness in alkaline media. Stability constant data shows that preferential chelation is similar to that of Versene and takes place in the following manner: $Cu^{+2}>Ni^{+2}>Pb^{+2}>Co^{+2}$, $Zn^{+2}>Cd^{+2}>Mn^{+2}>Ca^{+2}>Mg^{+2}$.

VERSENE-EDG

This is the disodium salt of N-β-hydroxyethyliminodiaceticacid. Its chemical structure is as follows:

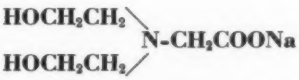


Except for the ferric chelates, this amino acid forms complexes with acid-to-metal ratios of 1:1 and 2:1 in the presence of heavy divalent metal ions but their stabilities are slightly lower than those formed with Versene-NTA. Yet, the general order of stabilities is the same. Actually, Versene-EDG is a weak chelant for the alkaline earth metals. In the presence of ferric ions it forms chelates having the empirical formula FeA and Fe_2A_3.

The ferric-Versene-EDG chelates are subject to some hydrolysis but are considerably more stable than those of Versene-NTA or Versene regular. This means that *Versene-EDG is an effective chelating agent for iron in alkaline solution.* At pH's below 10 it is a more effective chelating agent for ferric iron than Versene Fe-3 Specific. Above pH 10 Versene Fe3 Specific is more effective.

VERSENE Fe-3 SPECIFIC

This is the sodium salt of N,N-Dihydroxethylaminoacetic acid.



Versene Fe-3 Specific is *specifically* for chelating trivalent iron in alkaline solutions. It complexes other heavy metal ions but not as well as Versene-NTA or Versene-EDG. Nor does it chelate calcium or magnesium to any great degree.

Versene Fe-3 Specific forms complexes having acid-to-metal ratios of 1:1 (FeA) and 3:2 (Fe_2A_3) chelates in the presence of ferric iron. These are stable against hydrolysis even in strongly caustic solutions (pH≅ 13). It is the most efficient of the Versenes for iron chelation at measureable pH's above 10.

Because these new chelants make possible the control of metal ions at various levels of activity, they are invaluable tools of research. They should aid in the separation of metals, in the purification of alkaline earth metals, in the purification of fats, oils and other organic chemicals and also in inhibiting the activity of pro-oxidant metals. All are worthy of investigation. Samples of Versene-NTA, Versene-EDG Acid and Versene-EDG Sodium Salt (commercial grade) as well as Versene Fe3 Specific are available on request. Write Dept. L.

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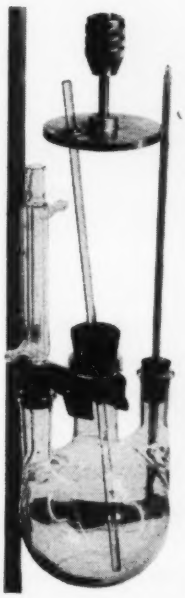

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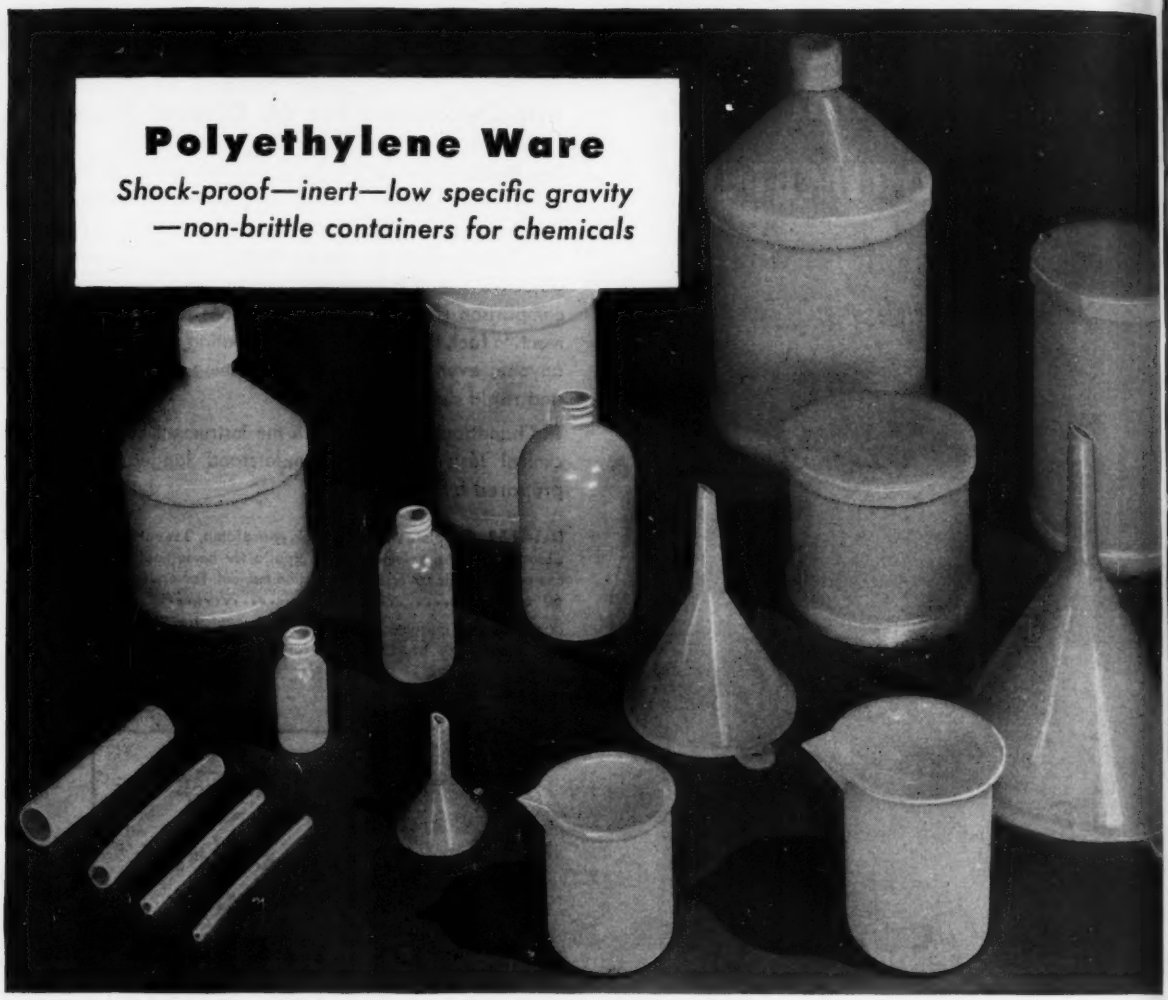

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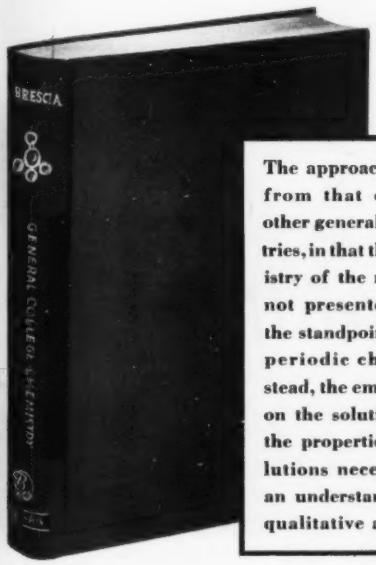

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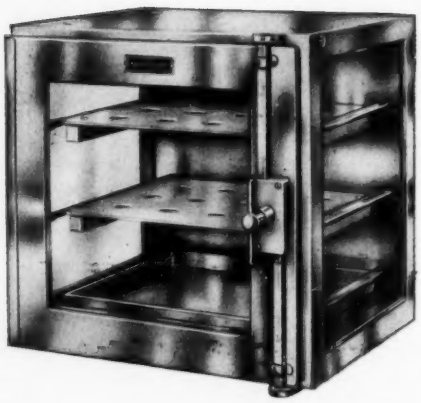

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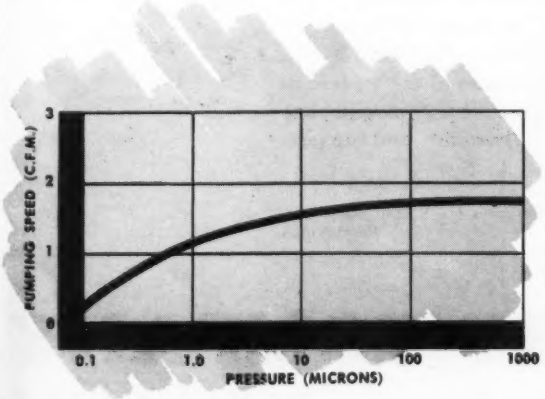

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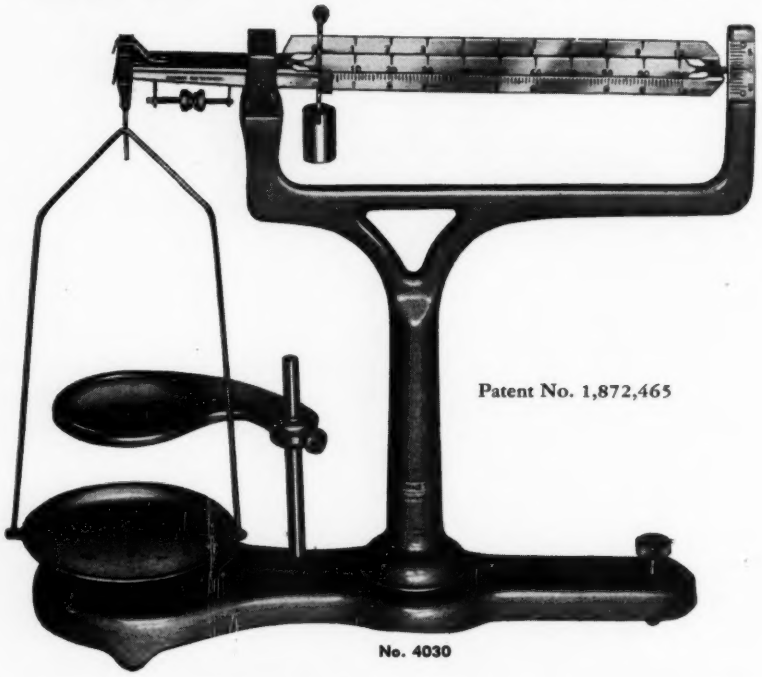

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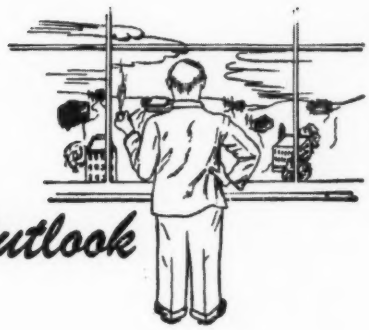


Editor's Outlook

Most of us agree that the advantages of the metric system are great; we might be glad to see its popular adoption. Dr. George B. Pegram, of Columbia University, recently pointed out one thing about it, however, which is interesting to consider.[1] He says:

> The designers of the metric system applied their knowledge of Greek and Latin very cleverly to making a lot of long new words that explain themselves immediately to persons who have even a slight knowledge of etymology, but they sadly neglected to consider the practicable usability of these words and the repugnance often felt by busy men who do not reflect on the reason toward quadrisyllabic terminology. . . . How long would we stand for calling a dime a "decidollar" or a cent a "centidollar?" A "decidollar" would very quickly become a "dess" and "cent" was fore-ordained. Again, suppose a foot were a tenth of a yard, an inch a tenth of a foot, and a mile just a thousand yards, thus realizing the advantage of a decimal system. How would we like to call these units a "yarder," a "deciyarder," a "centiyarder," a "kiloyarder?" Who would care to say, "The car flashed by at two kiloyarders a minute?"

[1] Pegram, G. B., "Units and Names," *Trans. N. Y. Acad. Sci.*, (II) **15**, 71 (1953).

The words which we use for our common units of measurement should not be clumsier than the good little four-letter words which we use so often. How about agitating, therefore, for the common use of "cen," rather than centimeter; "mim," rather than millimeter; "mil" or "eml," rather than milliliter. In fact, perhaps "meter" itself should be contracted to "mete" (which would at least distract us from the inconsistent pronunciation of "kilometer"). Pegram also thinks we could well shorten some of our other units to such terms as "sec," "coul," "frad," "hen," and "mig." I would add my own ten-cents-worth and suggest "grat" (for gram-atom), and "mygrat" (for microgram-atom). We have precedent in the "mol," originally a contraction from gram-molecule, but later (very handily) extended to apply to any gram-formula-weight.

The question of how to accomplish such desirable reforms is perhaps difficult. But Pegram thinks that something might be accomplished via such international organizations as UNESCO, itself an example of the desirability of indicating what we mean without a lot of words.

ASPECTS OF ISOMERISM AND MESOMERISM

I. (a) Formulas and Their Meaning (b) Mesomerism

RICHARD L. BENT
Eastman Kodak Company, Rochester, New York

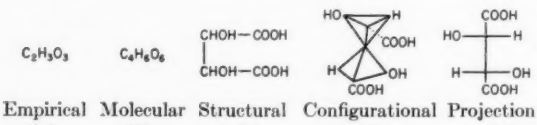
It is the object of the present series of articles to review the fundamental concepts of both mesomerism and isomerism, although the major part of the discussion is concerned with the latter. Notwithstanding the considerable number of texts devoted to these subjects, certain concepts continue to be misunderstood by many chemists. Particular care has been taken in these articles to give satisfactory definitions of the terms used, because frequently a poor definition can contaminate the whole of one's thinking concerning a subject.

For a proper understanding of either isomerism or mesomerism, it is important to have a clear idea as to the *exact* meaning and limitations of the various types of formulas that are commonly employed. For this reason, a fairly detailed discussion of each type is given in the first part of the present paper. A consideration of the structural formula inevitably brings up the question of resonance (mesomerism), and a discussion of the latter constitutes the remainder of Part I. Part II will be concerned with structural isomerism and Part III with stereoisomerism, the discussion throughout being limited almost entirely to compounds of carbon.

It should be emphasized from the start that the concepts and principles of isomerism are fundamentally different from those of mesomerism. Isomerism is concerned with the arrangement of *atoms* in a molecule (or ion), while the theory of resonance (in organic chemistry) is concerned with the bonds linking those atoms, *i. e.*, with *electrons*. Nevertheless, the theory of resonance does enable one to state more precisely just what the theory of isomerism can and cannot tell us.

FORMULAS AND THEIR MEANING

Let us consider, for example, the following various ways in which one of the optically active forms of tartaric acid can be represented:
The formulas contain additional information as one proceeds from I*a* to I*d*, the spatial or configurational



Empirical (*a*) Molecular (*b*) Structural (*c*) Configurational (*d*) Projection (*e*)

I

formula being the most complicated. The so-called projection formula I*e*, however, is merely a planar (and therefore simplified) representation of the configurational formula. Let us now consider in detail the meaning of each of these formulas (without being concerned with experimental details).

MOLECULAR AND EMPIRICAL FORMULAS

In order to make our definitions more precise, let us first consider the *covalent* substances. The *molecular* formula of a substance (element or compound) not only tells us which elements are present in a molecule of the substance but exactly how many atoms of each element. Some covalent substances, however, cannot be said to possess molecular formulas because they do not consist of discrete molecules. Thus the element carbon, which cannot be liquefied, exists in its various crystalline forms as "giant molecules." In diamond the covalent bonds between the carbon atoms extend throughout the whole crystal, while in graphite the covalent bonds extend in the plane of the hexagonal layers of carbon atoms (the layers themselves being held together by van der Waals forces).[1]

The *empirical* or *proportional* formula of a substance (obtained directly from its percentage composition) tells us the proportions of the elements that are present. Empirical formulas (in contrast to molecular) can be assigned to all pure substances. Because it is possible to represent the proportions of the elements in a compound in a number of different ways, *i. e.*, by a number of different formulas, in practice, the empirical formula is defined as the simplest proportional formula capable of representing the substance. For those substances consisting of molecules, it expresses the *relative* numbers of atoms present in a molecule of the substance in the smallest whole numbers. Thus, although $C_1H_{1.5}O_{1.5}$, for example, is an accurate proportional formula for tartaric acid, it is not normally called the empirical formula, because fractional terms (*i. e.*,

[1] See Pauling, L., "The Nature of the Chemical Bond," 2nd ed., Cornell University Press, Ithaca, New York, **1940**, p. 172.

fractions of an atom) obviously cannot be present.

The empirical formula for tartaric acid, $C_2H_3O_3$, informs us that for every two atoms of carbon that are present in the molecule (whatever the actual number is) there also have to be three atoms of hydrogen and three atoms of oxygen. Thus its molecular formula can be represented by the expression $(C_2H_3O_3)_n$, where n may be 1, 2, 3, or any other whole number. From the experimental value for the molecular weight of tartaric acid it is found that $n = 2$, *i. e.*, the molecular formula of tartaric acid is $(C_2H_3O_3)_2$ or $C_4H_6O_6$. In many instances, of course, the molecular formula of a compound is identical with its empirical formula.

Surprisingly enough, there is a certain inconsistency in the usage of empirical and molecular formulas, particularly in representing polymers. Thus, considering an addition polymer, the empirical formula of the rubber obtained by the polymerization of isoprene is usually written as $(C_5H_8)_n$, where n represents an unknown whole number. According to the usual definition of an empirical formula, the rubber should be represented by the formula, C_5H_8, since this represents the simplest proportional formula; the expression $(C_5H_8)_n$ would be regarded as the molecular formula, even though the actual number of monomeric units might not be known. With most high polymers, of course, the number of monomeric units per molecule is not constant and may vary within fairly wide limits.

The $(X)_n$ formulation for the empirical formula is also generally used for condensation polymers, though sometimes incorrectly. The empirical formula of starch, for example, is almost invariably written as $(C_6H_{10}O_5)_n$. The correct formula, however, is $(C_6H_{10}O_5)_n$ HOH, for the latter at the same time represents the disaccharides, trisaccharides, etc., which it should.[2] For example, sucrose formed by the condensation of two glucose molecules,

$$H—C_6H_{10}O_5—\boxed{OH\ H}—C_6H_{10}O_5—OH$$

possesses a *molecular formula*, $C_{12}H_{22}O_{11}$. Obviously, the expression $(C_6H_{10}O_5)_n$ cannot represent its empirical formula.[3]

Let us now consider the ionic compounds. Although empirical formulas can be assigned to them, they cannot be said to possess molecular formulas since they are composed of ions rather than molecules. Nevertheless, in practice, particularly in regard to organic compounds, formulas which are in many respects equivalent to molecular formulas are assigned to the ionic compounds. Thus, disodium tartrate, for example, is assigned the formula $C_4H_4O_6Na_2$ or $[C_4H_4O_6]^{--}[Na^+]_2$. Although these are obviously different from the empirical formula of the substance, $C_2H_2O_3Na$, they are frequently called simply "formulas" without further designation. To avoid ambiguity, it is preferable to call such a formula an *ionic formula*, and it tells us how many atoms (charged or uncharged) of each element are present in the smallest group of ions capable of representing the actual properties of the substance.

An ionic formula is perhaps a somewhat "artificial" concept since it represents an isolated group of ions. Such a group is not normally capable of independent existence in the presence of other ions. Thus, in the crystalline state, the attractive forces extend from ion to ion throughout the crystal, while even in solution the ions are not completely independent of one another. The group of atoms in a neutral molecule, of course, is normally capable of independent existence.

Whatever the theoretical objections to the concept of an ionic formula, the fact remains that it is an extremely useful concept. While it is true that many of the simpler ionic compounds (particularly inorganic) are adequately represented by their empirical formulas (because they are often identical with their ionic formulas), this is not true in the case of the organic ionic compounds. In fact, the organic chemist not only wants to know how many atoms of each element are present, but also the structural and even spatial arrangements of the atoms in the respective ions. The very concept of an ionic formula enables him to assign structural and configurational formulas to the ionic compounds in much the same way as for the strictly covalently linked compounds. In addition it enables one to include such compounds into the normal framework of definitions in isomerism without much difficulty. Thus, while butyric and isobutyric acids, for example, are said to be isomeric because they possess identical molecular formulas, their sodium salts can be called isomeric because they possess identical ionic formulas.[4]

THE STRUCTURAL FORMULA

In regard to covalent substances, the (classical) structural, or constitutional, formula tells us how many atoms of each element are present in a molecule of the substance, *which atom is linked to which*, and in each case whether the linkages are single, double, or triple. However, a "modern" structural formula contains further information, as will be shown.

In some ways the term "structural" as applied to a formula is misleading, for it is easy to attach to the word structure a three-dimensional or spatial meaning. This is due firstly to our everyday use of the word, as when we talk about the structure of a table or building, in which something with three dimensions is implied;

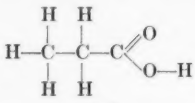
[2] As pointed out by WRIGHT, K. E., J. CHEM. EDUC., **24, 562** (1947).

[3] Starch consists of such a large number of condensed glucose units (or rather it can be considered to consist of condensed glucose units since the exact mechanism by which starch is formed is not yet known) that the difference between $(C_6H_{10}O_5)_n$ and $(C_6H_{10}O_5)_n$ HOH could not, of course, be detected analytically. Strictly speaking, because of "irregularities" in regard to end groups which occur, for example, in starch and, in fact, in all high polymers, only "approximate" formulas can really be assigned to such compounds.

[4] Unfortunately, there is no agreement in the standard texts as to exactly what compounds should or should not be called isomeric, and therefore the basic definitions differ somewhat (at least in regard to ionic compounds). The definitions given above are preferred by the present writer because of their inherent simplicity (see Part II for further discussion).

secondly, having known for a long time about the spatial aspects of a molecule, we are prone to see more in a structural formula than is really there. Although a structural formula is sometimes called a planar formula, it is best described as being a "noncommital" formula, since it does not tell us *anything* about the spatial relationships of the atoms. It is true of course, that *on paper* structural formulas are represented as being in the plane of the paper, but this is purely for convenience and does *not* mean that the distribution of atoms is to be regarded as being planar. A structural formula will tell us, for example, that atom A is linked to atom B, and that B is linked to C, and so on, but it does not tell us whether these atoms are joined together in a straight line or what their spatial positions are. It required the tetrahedral concept of the carbon atom, by Le Bel and van't Hoff, to give us the additional knowledge, as will be shown in the section on stereochemistry (Part III).

In practice, the structural formula of a compound is written in a number of different ways, but it must be carefully noted that they all possess exactly the same meaning. The structural formula of propionic acid, for example, can be written in any of the following ways.



(*a*)

$CH_3—CH_2—COOH$

(*b*)

CH_3CH_2COOH

(*c*)

C_2H_5COOH

(*d*)

II

Formula II*a*, in which every individual atom and valency is represented separately, is sometimes called a *graphic* formula. This method of writing out the structural formula in full, although giving us a vivid picture as to the way in which the atoms are linked up, is not often employed. Instead, the simpler formulas, II*b* to *d*, are often used, since the latter are more easily written and occupy less space. Nevertheless, Formulas (*b*) to (*d*) imply (*a*) and are merely examples of chemical shorthand.

In practice, an ionic compound such as sodium ethoxide, for example, is conveniently assigned the structural formula CH_3CH_2ONa, or (preferably) $[CH_3CH_2O]^-\ [Na]^+$. Strictly speaking the latter represents the structural formula of an isolated pair of ions only (see earlier remarks concerning ionic formulas).

In order to include a number of compounds which would otherwise be incapable of representation by structural formulas, certain legitimate extensions to the original meaning of the latter have been made. Because each covalent bond is now known to represent a shared pair of electrons, a simple free radical like methyl is assigned the structural formula $\cdot CH_3$, the single dot implying one unshared electron.

A semipolar bond is now included as part of a "modern" structural formula. Because both of the shared electrons in a semipolar bond come from one atom (the donor), the latter possesses a formal positive charge, while the acceptor possesses a formal negative charge. An amine oxide, for example, is represented by III*a*. An alternative formulation is shown by III*b*. Both representations are, of course, identical in meaning.

$R_3N \rightarrow O$ (*a*) $R_3N^{(+)}—O^{(-)}$ (*b*)

III

The situation is not entirely clear as regards the corresponding sulfur compounds. Thus the sulfoxides have been represented for a number of years by IV. However, in contrast to the elements of the first period, sulfur is capable of expanding its valency shell and hence a formula such as V is possible.

$R_2S \rightarrow O$ IV $R_2S{=}O$ V

The present attitude, based on considerable experimental evidence, is that the bond between the sulfur and oxygen (in such a compound) is neither a semipolar bond nor a double bond, but is *intermediate* between the two. This means that a sulfoxide cannot be represented by a single structural formula. The actual state of the molecule has to be represented as a *resonance hybrid*, although there still seems to be considerable doubt concerning the extent to which each of these forms contributes to the hybrid.[5]

Another example of a resonance hybrid is the triphenylmethyl radical, which also cannot be represented by any one structural formula. For convenience, its "structure" is often written $(C_6H_5)_3C\cdot$, as if the unshared electron were attached to the acyclic carbon atom. The electron does not remain there, however, and is part of the time located on the carbon atoms of the benzene rings. A detailed explanation of such mesomeric (resonance) systems is given later.

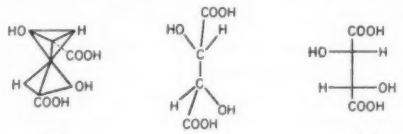
CONFIGURATIONAL AND PROJECTION FORMULAS

Since these formulas will also be considered in Part III of this series, only a brief account is given here. The stereochemical or configurational formula is a *spatial* formula. In regard to the covalent compounds, it tells us how many atoms of each element are present in a molecule of the substance, which atom is linked to which, and the *relative* spatial positions of the atoms. Although a configurational formula is often called a three-dimensional formula, the term spatial is preferable. The majority of molecules do, of course, extend in three dimensions and their respective configurational formulas will show this. However, configurational formulas also portray planar molecules (*e. g.*, ethylene) and even linear molecules (*e. g.*, acetylene), so it is not quite accurate to classify all of them as being three-dimensional.

According to stereochemical theory, a molecule such as tartaric acid, which can be regarded as a fully sub-

[5] See VOGEL, A. I., *et al.*, *J. Chem. Soc.*, **1952**, 535, and references given there.

stituted ethane, is represented by Formula VI*a*, *i. e.*, by a point-to-point union of two tetrahedra (one of the optically active forms is represented).



(*a*) (*b*) (*c*)

VI

The hydrogen, hydroxyl, and carboxyl groups are considered to be at the corners of the two tetrahedra and, in each case, the carbon atom to which these groups are attached is considered to be at the center of the corresponding tetrahedron. The "central" carbon atoms are often not shown when using this type of spatial formula.

An alternative way of picturing the molecule is by the valence-bond type of formula as shown by VI*b*. With this type, the location of both the bonds and atoms (or groups) in the molecule is apparent. The differences between the (three) isomeric tartaric acids (stereoisomeric, since they all possess the same structural formula) can be shown without drawing in full the spatial relationships that exist between the atoms in the hydroxyl and carboxyl groups. The spatial relationships of the atoms or groups about each asymmetric center[6] (in this case, each of the two central carbon atoms) constitute the really important thing to be represented. When there is no asymmetric center, as in the carboxyl group, for example, the ordinary structural formula contains enough information because such a group, by itself, is not capable of causing additional isomerism. Hence, in practice, we use a formula that is really only "partly" spatial, the "rest" of the formula being written out in the form of a structural formula. Nevertheless, such a formula is always called configurational.

It should be pointed out that even today it is not known definitely whether VI*a* actually represents the dextro or levo form of tartaric acid, although recently there have been claims in regard to the determination of the absolute configuration of such compounds. All that we can say for certainty is that if VI*a* does actually represent the dextro configuration, then the levo form *must* be represented by its mirror image, VII*a* (or vice versa).

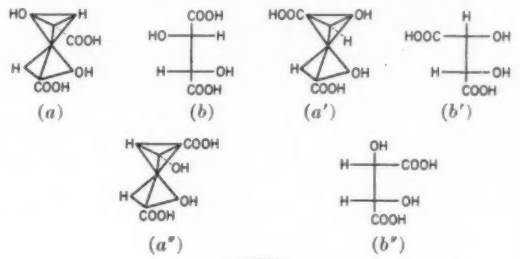
(*a*) (*b*)

VII

Because of the occurrence of freedom of rotation about the single bonds, each of the tartaric acid molecules (whatever their absolute configuration may be) can only be represented properly by an infinite number of spatial formulas. In practice, of course, one of these "static" formulas (representing an instantaneous form) is chosen to represent the dynamic molecule.

Owing to the tediousness of writing out configurational formulas of complicated molecules, a simpler method of representing them was devised, resulting in what is known as projection formulas. Thus, VI*a* and VII*a* are represented by the projection formulas, VI*c* and VII*b*, respectively. Certain rules must be carefully observed when using projection formulas. They may be moved around *only in the plane of the paper;* they cannot be lifted out of the plane and inverted, for if they are, they cease to represent the original molecule. Thus, if VI*c* is inverted, it becomes identical with VII*b*.

Let us examine the molecule represented by VI*a* more closely. Because of the relative freedom of rotation between the central carbon atoms, the molecule can also be represented by VIII*a*, *a'*, or *a''* (the counterclockwise relationship of H, OH, and COOH, taken in that order, being preserved) and, in fact, by many other formulas "in between" these, although the various spatial positions are not all equally probable.



(*a*) (*b*) (*a'*) (*b'*)

(*a''*) (*b''*)

VIII

Their corresponding projection formulas can be represented by VIII*b*, *b'*, or *b''*. It should be noticed that the atoms or groups situated at the top or bottom of the vertical line in projection formulas (in one commonly accepted system, and in this paper) represent those atoms or groups which project in front of the plane of the paper (in the other system, behind the plane). For convenience, one of these, VIII*a*, is arbitrarily chosen to be the correct formula. Similarly, with the other isomeric tartaric acids, the formulas are so chosen that the carboxyl groups always project in front of the plane of the paper.

Notwithstanding the above remarks, a projection formula (rather than a configurational one) is often chosen arbitrarily to represent a molecule. First of all, projection formulas are generally used instead of the more complicated spatial formulas, and secondly *absolute* configurations of many optically active molecules are not known. Thus, VI*c*, for example, has been chosen (by universal agreement) to represent *levo* tartaric acid, *it being understood that it is uncertain whether this actually* represents VI*a* or its mirror image.

[6] A carbon atom attached to four different atoms or groups is said to be asymmetric. The tartaric acid molecule contains two *similar* asymmetric carbon atoms and is therefore capable of existing in a dextro, levo, and meso form (see Part III).

MESOMERISM

The term "electronic isomer" has been used occasionally in the past to describe structures that differ only in the position of *electrons*, *e. g.*, the two Kekulé structures for benzene. The expression is completely misleading, however, for although one can (and does for reasons of convenience) write down such electronic structures on paper, in principle they do not exist as separate entities. A molecule like benzene does not possess one electronic structure part of the time and another structure (or structures) the rest of the time. The actual electronic state of the molecule all of the time is "in between" the possible structures (there is an intermediate density of electric charge between any two carbon atoms). The phenomenon was appropriately called *mesomerism* by Ingold (the word meaning literally "between the parts"). Another way of describing the mesomeric state of a molecule is to state that it is in *resonance* among a number of electronic structures. Thus, benzene is said to resonate between the two Kekulé structures. It is unfortunate that the term resonance is likely to give one the erroneous idea that some kind of a "mechanical" oscillation is involved. It should be remembered, however, that the terminology adopted is extremely useful, for otherwise we would have to dispense with many of the classical structural formulas. Thus, although one is unable to write down on paper a classical formula for the mesomeric state of a molecule, one can describe it in terms of a number of structures that can be written classically.

This does not mean that such a molecule cannot be represented in some other manner. Thus, the *molecular orbital*[7] treatment of resonance makes use of a radically different type of formula. In fact, the latter (in contrast to the valence-bond treatment) attempts to give an actual "picture" of the mesomeric state of a molecule. The molecular orbital method is admittedly superior in a number of respects. However, because the latter does not make use of the classical structural formulas, the valence-bond method will be adhered to in the present paper.

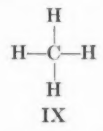
It might be pointed out that the theory of resonance is also concerned with certain phenomena in physics. Thus, for example, the spectrum of the helium atom can be explained only by assuming that its two electrons affect one another in such a manner that a so-called *exchange* takes place. The two electrons are said to be in resonance. In fact, the term resonance was first used in this connection. Here again, the phenomenon is not to be represented as a simple "mechanical" interchange.

Although the terms resonance and mesomerism are often used interchangeably in organic chemistry, it should be noted that the meaning of the term mesomerism is also frequently restricted, it being regarded as a special branch of resonance.

For classification purposes, one can state that the theory of resonance in organic chemistry is concerned with the following phenomena: (1) the formation of a *covalent* bond (or bonds) between identical atoms; (2) the formation of such a bond (or bonds) between dissimilar atoms; (3) the fact that it is possible to write for certain types of molecules (*e. g.*, benzene), ions (*e. g.*, the carboxylic acid ion), or free radicals (*e. g.*, the triphenylmethyl radical), two or more possible electronic structures. The actual state of such a molecule (ion or radical) is considered to be represented not by any one of those structures, but by all of them.

Resonance of type (3) which occurs between different valence-bond structures in certain compounds possessing multiple bonds (and frequently unshared pairs of electrons as well), is often called mesomerism. As mentioned above, however, the terms resonance and mesomerism are frequently used more or less interchangeably. Presumably this is because all resonating systems (and not just type (3)) can really be represented by some kind of "intermediate" electronic state which might therefore be termed mesomeric.

Probably the reason for sometimes limiting the term mesomerism to include only resonance of type (3) is the fact that once the fundamental nature of the covalent bond has been understood, the chemist can often write his formulas in the classical manner without further consideration. Thus, most of the compounds containing only single bonds are adequately represented (in many of their aspects) by the classical structural formulas. For example, the structural formula for methane, IX, accurately describes many of the relevant



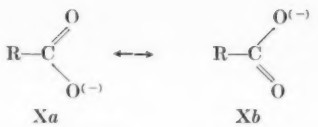
IX

properties of that compound. The chemist can, so to speak, "forget" that resonance is actually responsible for the formation of each of the covalent bonds.

A considerable number of compounds containing multiple bonds can also be represented quite accurately by the classical formulas, *e. g.*, ethylene, acetylene, etc. Nevertheless there are many compounds containing multiple bonds, particularly those with conjugated systems, that cannot be represented by classical structural formulas. For, in these compounds, *additional* resonance occurs, over and above the resonance that is responsible for each individual bond, and the resonance is of a type that completely invalidates the classical formulation.

Thus, the carboxylate ion is not represented either by X*a* or X*b*. Both formulas imply that one oxygen atom is doubled, linked and uncharged and that the

[7] For an elementary discussion of it, see NOLLER, C. R., J. CHEM. EDUC., **27**, 504 (1950). For a more advanced treatment, see COULSON, C. A., "Valence," Oxford University Press, London, 1952.



Xa Xb

other oxygen is singly linked and negatively charged. The ion is said to resonate between the two structures, meaning that the actual state of the molecule is all of the time "intermediate" between the two structures. The resulting electronic state of the molecule is also called a *resonance hybrid* to emphasize that a new type of structure has been formed. The electronic charge distributes itself equally between both oxygens, which thus receive a partial charge only. It is true, of course, that if a positive ion approached closely enough to one of the oxygen atoms, the latter would become predominantly, if not wholly, charged. In fact, this transference of charge or electrons from one atom to another is often very important at the "instant" a chemical reaction takes place.

It should be noted carefully that the term "intermediate" does not mean that the properties of the molecule are simply an arithmetical average of the properties of the individual structures, for resonance profoundly affects many of the properties of a molecule. Thus, the stability of the carboxylate ion is increased by the resonance, and the bond distances are shortened; the carbon-oxygen bond distance is not halfway between that of a single bond and a double bond but is actually closer to that of a double bond.

The general theory of mesomerism, or resonance, should be distinguished from the so-called *mesomeric effect*;[8] the latter, although a resonance phenomenon of type (3) is exhibited by a special class of compounds only. Without giving a rigid definition of the mesomeric effect, it can be stated that, in general, it is concerned with the tendency of an atom possessing an *unshared pair* of electrons (*e. g.*, nitrogen or oxygen) to enter into the resonance phenomenon when it is connected to a conjugated system of bonds. Thus, while the Kekulé forms of benzene exemplify the resonance phenomenon, type (3), benzene is not an example of the mesomeric effect, since no unshared pairs of electrons are involved in the resonance. A fairly detailed discussion of each type of resonance will now be given.

RESONANCE OF TYPE (1)

Let us consider the formation of a single covalent bond as exemplified in the hydrogen molecule. The theory of quantum-mechanical resonance, to use its full and somewhat terrifying name, was employed by Heitler and London in 1927 to explain the nature of the covalent bond. They postulated that the two electrons forming such a bond (which are said to be shared) take part in an exchange, or resonance, phenomenon somewhat similar to that which occurs in the helium atom.

If two isolated hydrogen atoms approach one another closely enough, and if the two electrons have *opposed* spins (this cannot be elaborated upon here), the following state of affairs occurs: The individual electrons no longer rotate solely about their respective nuclei but rotate about *both* nuclei.[9] This is expressed crudely by stating that the hydrogen molecule resonates between the two electronic structures, XI*a* and XI*b* (the letters refer to

$$\underset{\text{XI}a}{H_a^1\,H_b^2} \longleftrightarrow \underset{\text{XI}b}{H_a^2\,H_b^1}$$

the individual hydrogen nuclei and the numerals to the individual electrons). This symbolism merely means that after combination neither electron (1 or 2) belongs exclusively to either of the nuclei (*a* or *b*), but that both electrons are *mutually* owned by both nuclei. A mathematical treatment of the problem has shown that the electrons, while moving about both nuclei, spend the larger part of the time *between* the nuclei, and so we can think of the two positive nuclei as being held together by the "partially centralized" negative electrons by electrostatic forces ("partially" because they are in constant motion about the two nuclei). Unfortunately, the complete explanation as to *why* the electrons move in such a manner is extremely complicated and can be understood by the mathematical physicist only. This is because the theory of resonance is based on the highly mathematical concepts of quantum mechanics (or wave mechanics). The latter subjects are, of course, completely outside the scope of the present article, and so a few brief (and oversimplified) remarks will have to suffice.

Matter (as well as electromagnetic radiation) has a dual nature; it possesses both wavelike and particle-like characteristics. It is only when atomic dimensions are involved, however, that the wave nature of matter begins to make itself felt. The wave-mechanical picture of the hydrogen atom is that the electron, in its motion about the nucleus, is "guided" or "directed" by an accompanying system of waves (electron waves). In addition, these waves control the energy possessed by the electron (the waves themselves are not considered to possess any energy).[10]

When two hydrogen atoms approach one another closely enough, the two individual wave systems "combine" and a *new* wave system is set up which

[8] For a discussion of it, see JOHNSON, J. R., in GILMAN'S "Organic Chemistry," 2nd ed., John Wiley & Sons, Inc., New York, 1943, Vol. 2, pp. 1821–942; also REMICK, A. E., "Electronic Interpretations of Organic Chemistry," 2nd ed., John Wiley & Sons, Inc., New York, 1949.

[9] The two atoms will not combine, however, unless they can get rid of their excess energy, *i. e.*, it will normally require the presence of other particles to carry off the excess energy.

[10] For a lucid discussion of the nature of these waves (which are called "probability" waves), see JEANS, J., "Physics and Philosophy," The Macmillan Co., New York, 1945, pp. 123–71; see also BORN, M., "The Restless Universe," 2nd ed., Dover Publications, Inc., New York, 1951, pp. 106–65; MARGENAU, H., "The Nature of Physical Reality," McGraw-Hill Book Co., Inc., New York, 1950, pp. 307–88; WISWESSER, W. J., J. CHEM. EDUC., 22, 314, 370, 418 (1945).

(it can be shown) causes the electrons to possess *less* energy than they had before combination. The excess energy is given out as heat and is called the resonance energy of the molecule. The latter is a measure of the strength or stability of the covalent bond in hydrogen, since that amount of energy must be put into the molecule to break the bond.

The actual forces involved in the molecule, once the bond has been formed, are still the electrical forces of attraction between protons and electrons. The two hydrogen atoms combine, however, not because of any "immediate" electrical attraction, but because of the wave nature of the electron, which, so to speak, forces them to assume, on the average, "positions" of attraction; *i. e.*, the electrons spend the large part of the time *between* the nuclei. Although, in addition, mutual polarization occurs between the two hydrogen atoms, resulting in a small dipolar attraction, by itself it is not strong enough to account for the forces actually involved (the magnetic and gravitational forces are too small to have any appreciable influence).[11]

In general, it can be stated that when resonance occurs in a molecule (of whatever type) it appears to give more "freedom of movement" to the electrons. Thus, in the case of the hydrogen molecule (and similarly for all covalent bonds), the electrons have more freedom after combination since they move about *two* nuclei instead of only one. As shown by quantum mechanics, the increased freedom results in the electrons possessing *less* energy than they would have without the resonance.

RESONANCE OF TYPE (2)[12]

The carbon-chlorine bond can be taken as an example of this type or resonance. The chlorine atom is considerably more electronegative than carbon, and it attracts the shared electron pair to a greater extent. The chlorine atom thus acquires a fractional negative charge and the carbon a fractional positive charge, the bond as a whole possessing a dipole moment. Diagrammatically, one can represent this state of affairs in either of two ways, by XII*a* or XII*b*. This manner of representation, however, does not tell the

C :Cl	$C^{\delta+} - Cl^{\delta-}$
XII*a*	XII*b*

whole story. There is another and more accurate way of looking at the problem. The bond is to be thought of as in resonance *primarily* between a covalent extreme, XIII*a*, and an ionic extreme, XIII*b*. The

C:Cl $\longleftrightarrow$	$C^{(+)}\ Cl^{(-)}$
XIII*a*	XIII*b*

actual electronic state of the bond is somewhere between these two extreme states, the two atoms acquiring fractional charges, as stated above. The important thing to realize, however, is that the actual strength of the intermediate, or hybrid, bond is not the arithmetical average of the bond strengths for the covalent and ionic extremes. The hybrid bond is actually stronger than either the covalent or ionic bond taken individually. It requires more energy to break the hybrid bond than would be necessary if the additional resonance did not occur (and by additional is meant the resonance over and above that which occurs in a "pure" covalent bond; a pure ionic bond, of course, is not a resonating system).

The greater the ionic contribution to the actual state of the bond, the greater will be the resonance, and the greater the strength of the bond. If the ionic contribution to a bond were exactly equal to the covalent contribution, the resonance interaction and the resultant strength of the bond would be at a maximum. In fact, if three (or more) structures of equal probability and stability were possible, then the strength of the hybrid bond would be even greater. If the contributions of the covalent and ionic extremes are not equal (as in the carbon-chlorine bond), then the resonance interaction will be less than the maximum theoretically possible, and the electronic state (and strength) of the hybrid bond will be closer to that possessed by the most stable structure. A single bond may lie anywhere between the ionic extreme and the covalent extreme, depending on the relative electronegativities of the atoms involved. Thus, in the series HF, HCl, HBr, and HI, the ionic character of the bond is greatest for the hydrogen fluoride and least for the hydrogen iodide.

RESONANCE OF TYPE (3)

As mentioned previously, a molecule like benzene cannot be described accurately even in terms of more than one structural formula, although we still attempt to so describe it. It should be noted that an accurate description of all *tautomeric* systems can be made in terms of two or more structures. At the equilibrium point, the following type of statement could be made which would apply to all tautomeric systems: The number of molecules possessing Structure A, the number possessing Structure B, and so on (depending upon the number of individual structures involved) will remain constant. Wording it somewhat differently, one could state that, over a period of time, on the average each molecule will spend a definite fraction of that time as Structure A, another definite fraction as Structure B, and so on, the individual periods of time remaining constant at equilibrium.

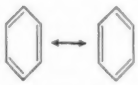
[11] For a more detailed (but nonmathematical) discussion of the covalent bond, see SLATER, J. C., "Introduction to Chemical Physics," McGraw-Hill Book Co., Inc., New York, **1939**, pp. 352–76; PAULING, L., ref. 1, pp. 3–24.

For interesting discussions, with particular emphasis on the theory of valency, see PALMER, W. G., "Valency, Classical and Modern," Cambridge University Press, Cambridge, England, **1946**; also, RÂY, P., "Theory of Valency and the Structure of Chemical Compounds," University College of Science, Calcutta, India, **1946**; also, COULSON, C. A., ref. 7.

For a more advanced treatment, see SYRKIN, Y. K., AND M. E. DYATKINA, "Structure of Molecules and the Chemical Bond," Interscience Publishers, Inc., New York, **1950**.

[12] For a more thorough discussion, see PAULING, L., ref. 1, pp. 34–75.

The simple concept of a "mechanical mixture" can accurately describe a tautomeric system. However, when the movements of electrons rather than atoms are involved, as in all resonating systems, the concept of a mechanical mixture is completely erroneous. When two structures differ only in "bond arrangements" rather than atom arrangements, the classical idea of an equilibrium has to be revised. Although no one classical structural formula (of the normal valence-bond type) can represent the benzene molecule, for convenience, or rather because of the desire to keep the outward form of the classical terminology, we speak of the benzene molecule as resonating between the two Kekulé structures,



(neglecting other minor contributing structures). Nevertheless, benzene is emphatically not a dynamic "mixture" of the two forms.

Thus, the bond between any two carbon atoms in benzene is not part of the time single and part of the time double, but all of the time "in between" a single and a double bond; it is conveniently called a "one and a half" bond. Nevertheless, because of the resonance, it is actually closer to being a double bond as regards bond strength and distance. However, considering the characteristic lack of reactivity of benzene (it undergoes substitution reactions rather than addition reactions), it is closer to behaving like a single bond. The resonance brings about an increased stability which prevents the double bonds from acting in the normal manner.

The concept of a bond being in between a single and a double bond should cause no confusion if a covalent bond is pictured as being a "time-averaged" cloud of negative electricity (which is perfectly justified, owing to the tremendous speed of the electrons). The density of charge between any two atoms which are covalently linked can be regarded as a measure of the strength of the bond.[13]

An isolated carbon-carbon double bond is stronger than an isolated single bond (but not twice as strong), since it consists of two pairs of shared electrons, and thus the density of charge is also greater.[14]

A bond that is described as being "in between" a single and a double bond merely implies that the density of electric charge for the bond is in between the densities exhibited by isolated single and double bonds. In other words, in benzene the electrons do not move, so that part of the time, between any two carbon atoms, the density of charge is equal to that of a single bond and part of the time it is equal to that of a double bond. They move so that the density of charge is *all* of the time in between the two extremes (but not halfway between). To show this and to demonstrate the complete equality of all the carbon-carbon bonds in benzene, the "formula" for the latter is occasionally represented by XIV.[15]

XIV

The properties of the carbon-carbon bond in benzene are not halfway between those of a single bond and a double bond because of the gain in stability due to the resonance that occurs. The molecule is a completely changed molecule, and the change is not one involving a simple averaging of the properties of the two hypothetical Kekulé structures. When one states that the stability of benzene is increased by the resonance, it means that the molecule has a lower energy content than would be expected from classical considerations. The word stability, in connection with resonance phenomena, always refers to a low (or relatively low) energy content, and this does not necessarily imply a low chemical reactivity, although this often occurs (as in benzene).

Thus, one finds that more heat is evolved than expected from ordinary calculations based on heats of formation for all carbon-carbon bonds in benzene, *i. e.*, on adding the heats of formation for three isolated single bonds and for three isolated double bonds, the total heat which would be given out is less than is actually found experimentally for the bonds in benzene. This means that more heat has to be put into the molecule to break the carbon-carbon bonds than would be required in the absence of resonance. The *excess* heat energy that is given up during formation is called the resonance energy of benzene and is a measure of the increased stability of the molecule.[16] When one speaks of the resonance energy of benzene, one is really referring to *additional* resonance energy—over and above

[13] Ionic bonds, of course, have no density of *charge*, but only "electric-field" density. Neither are we considering here a bond which is a resonance hybrid between an ionic bond and a covalent bond, *i. e.*, a bond with partial ionic character. In actual fact, there is no such thing as a "pure" covalent bond, but here we can consider that such a bond exists.

[14] Although a double bond is the stronger bond—and this means that more energy is required to break both of the bonds than to break a single bond—it is chemically more reactive. The second bond (which is called a π bond) is actually weaker than the first, and it is this bond that is preferentially attacked by chemical reagents. Thus, for example, it is more easily polarized by an attacking ion.

[15] It is interesting to note, however, that such a formula (originally proposed by the classical theory of partial valency) is now being used to represent a "simplified" *molecular-orbital* formula for benzene. According to molecular-orbital theory, all of the "secondary" bonds in benzene are equivalent because of a mutual overlapping of six p atomic orbitals (see ref. 7). Other mesomeric molecules can also be represented by such "dotted-line" formulas. For a text making extensive use of such formulas, see DEWAR, M. J. S., "The Electronic Theory of Organic Chemistry," Oxford University Press, London, 1949.

[16] For a very interesting discussion of the energy relationships involved in resonance systems, see BROOKER, L. G. S., "Resonance and organic chemistry," in BURK, R. E., AND O. GRUMMITT's "Advances in Nuclear Chemistry and Theoretical Organic Chemistry," Interscience Publishers, Inc., New York, 1945, pp. 63–136.

that required for each individual bond—brought about by the resonance that occurs between the two Kekulé forms. In general, when one is considering resonance phenomena of type (3), the word "additional" is normally omitted, since the "covalent bond resonance" that also occurs is taken for granted.

The resonance that occurs in benzene is said to be "complete," because the two contributing forms are identical. If the two forms were very different in electronic structure and stability, then very little resonance would occur, and the resultant molecule would not gain appreciably in stability but would closely approximate the electronic structure and stability of the more stable of the two forms. These remarks, of course, apply equally well to any molecule which can be considered to be a resonance hybrid.

ACKNOWLEDGMENT

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HERMANN MOHLER

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HERMANN MOHLER, who is now serving as the temporary head of the College of Arts and Sciences at the University of Bagdad, where he lectures in English on physical, industrial, and biochemistry, is in a sense returning the great service rendered to Europe by the Moors who brought Islamic science into Spain many centuries ago. Dr. Mohler, who is distinguished by his humanitarian outlook, is working in Iraq under the auspices of UNESCO.[1]

Born on March 2, 1900, at Liestal, Switzerland, he was graduated *magna cum laude* from the University of Basel where he took his doctorate (1925) with a major in physical chemistry and minors in botany and general chemistry. He early showed a strong interest in scientific problems and his two-year activity as assistant in the physical chemistry laboratory of his *alma mater* revealed that he had a great ability for teaching.

A year after the doctorate examination he was appointed provincial Food Inspector, and in 1930 he passed the federal examination for food chemist. Shortly thereafter, he went to Zurich as adjunct to the cantonal chemist, and two years later was named city chemist of Zurich. Here he had an opportunity to develop his outstanding organizing abilities and also his scientific activities.

In January, 1937, he habilitated as Privatdozent at Basel, receiving the *venia legendi* in physical chemistry. The habilitation essay bore the title: "Diacetyl as butter aroma and its spectroscopy." The habilitation lecture dealt with the problem of the relation of chemistry to the new physics.

His lectures have always been well attended. They covered a wide variety of topics, *e. g.*, chemical warfare; explosives; the relation of light absorption to chemical constitution; dipole moments; sewage; potable waters. More than 90 papers testify to the comprehensiveness of his chemical activity. In the field of pure chemistry, he has specialized on spectroscopy and electron theory. He is especially interested in the relations between physical properties and chemical constitution. In 1947 he was appointed extraordinary professor of physical chemistry at Basel.

He has acted as consultant to the military authorities on such matters as the analysis of toxic smoke screens, the detoxification of areas exposed to war gases such as Yperite, and protection against gas attacks in general.

He has the following books to his credit: "Lösungsspektren" (1937); "Beziehungen der Chemie zum Weltbild der Physik" (1939); "Das Absorptionspektrum der chemischen Bindung" (1943), which was out of print within eight months and was reprinted in the U. S. A. in 1946; "Optische Methoden des Chemikers" (1945); "Elektronentheorie der Chemie" (1946); "Chemische Optik" (1950).

The Werder Foundation awarded him its Werder Medal in 1944 in recognition of his outstanding achievements in food chemistry and his method of analyzing spirituous liquors. Since 1946 he has served as president of the Schweizischer Chemiker-Verband and since 1947 he has been head of the editorial board of *Chimia*, which he founded.

[1] MOHLER, H., "Die Wissenschaft in Aufbauwerk der Unesco," *Chimia*, 6, 153 (1952).

THE SCOPE AND FUTURE OF ISOTOPE UTILIZATION[1]

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To appreciate the growth, and more particularly the future, of isotope utilization, we should go back almost 60 years to the discovery of radioactivity. From Becquerel's discovery in 1896 to the present, radioactivity has been the key that has opened up door after door in the dramatic development of nuclear science or atomic energy.

By 1900 five different radioactive elements were known; four years later this number was extended to 20; in 1912 it was 30; and today more than 40 naturally occurring radioactive species are known to exist. In 1911 radioactivity indirectly led to the concept of isotopes. By that time nearly 30 radioactive "elements" had been discovered, yet only 12 positions could be found in the periodic chart to accommodate them. Shortly thereafter, F. Soddy in presenting a paper on "Radioelements and the periodic law" before a meeting of the Chemistry Section of the British Association in Birmingham concluded "that all elements falling into the same place in the periodic table...are chemically identical and nonseparable by chemical methods." Later the same year Soddy proposed the name isotopes to describe such "elements." Thus, studies of radioactivity were the key to the concept of isotopes.

This concept, coupled with the fact that a stable element can have a radioactive isotope, led to the disclosure in 1913 by Hevesy and Paneth that a small amount of radioactive lead could be used as a radioactive tracer or indicator for lead in the determination of the solubilities of sparingly soluble lead salts. Radioactivity, therefore, was the key to the first tracer experiment.

In the years that followed, radioactivity brought forth a number of concepts which were to prove extremely important to the development of nuclear science. In 1912 Rutherford's interpretation of the large-angle scattering of alpha particles passing through thin sheets of metal led to the disclosure of the nucleus, or as Rutherford put it, "the atom contains a central charge distributed through a very small volume." Seven years later, working with the same alpha rays, Rutherford achieved the first controlled artificial disintegration of an atomic nucleus. In 1932 the discovery of the neutron was credited to Chadwick for his explanation of observations made by Bothe and Becker, the Joliots, and H. C. Webster when alpha rays from polonium were impinged on light elements, notably beryllium. In 1934 I. Joliot-Curie and her husband, F. Joliot, using a similar natural source for irradiations, discovered that radioactivity could be induced in stable elements. Here, then, the study and use of natural radioelements unexpectedly opened up the door to induced radioactivity in other elements.

At about the same time E. O. Lawrence of this country invented the cyclotron and it was quickly observed that this and other particle accelerators could be used to induce radioactivity in a large variety of elements. The use of such man-made radioactivity was, however, limited largely to laboratory research problems. Generally, only experiments requiring small amounts of radiomaterials could be performed, since the quantities which could be produced were limited and costly.

Then in 1939 came the discovery of fission. Here again radioactivity led the way. Fermi and his coworkers had observed in 1934, during their investigations of the nuclear reactions produced in uranium by neutrons, that at least four different beta activities, distinguished by their half lives, could be detected. Five years later Hahn showed these beta-ray emitters to be the fission fragments of uranium.

Then followed in rapid succession the discovery that uranium fission results in neutron production, that a chain reaction could be sustained, and that these two phenomena could serve as the basis for the design and operation of a nuclear reactor. With the advent of the reactor it became possible to produce radiomaterials in sufficient quantities to permit their wide-scale application in quantities both large and small. This opened up entirely new roles and new horizons for radioactivity.

Radioactivity was not only the key to numerous developments in atomic and nuclear science, but as a label or tag on atoms, namely, as a radioisotope, it has led to many new concepts in the physical and life sciences, particularly in biology. Undoubtedly there will be many other developments arising from the use of radioactivity in the years to come, but it is difficult to predict the future scope and developments of isotope utilization. Certain areas, however, are subject to reasonable forecast. It will be the intent of this paper to discuss the current extent and possible future scope of isotope utilization, to describe the necessary role of chemistry, and finally to indicate wherein the chemical educator may contribute to future developments.

[1] Presented at the 122nd Meeting of the American Chemical Society, Atlantic City, New Jersey, September 16, 1952.

CURRENT SCOPE

Since the start of the Atomic Energy Commission's isotope distribution program in August, 1946, more than 28,000 shipments of radioisotopes have been made from the production facilities in Oak Ridge, Tennessee, to some 1700 departments of nearly 1000 institutions throughout the United States. The shipments have gone to 46 states, the District of Columbia, and Hawaii.

In addition, over 5000 shipments have gone to other AEC installations and nearly 1500 shipments have gone to some 34 foreign countries. And the program is continuing to expand rapidly.

Added to the shipments from Oak Ridge are those from Brookhaven National Laboratory and Argonne National Laboratory. Also, more than 6000 shipments have been made from secondary commercial suppliers in the form of labeled compounds, sealed sources, and other specially prepared radiomaterials.

If we were to survey the scientific and technological fields in which radioisotopes have been used, we would find them employed in nearly every phase of biological and physical research, as well as in many of the applied problems of medicine, agriculture, and industry. Another measure of the nature and scope of radioisotope utilization may be gained by determining the number and types of laboratories using them.

A check of isotope shipments by field of use shows that only about four per cent of the total number of shipments have been used for chemical research. Although the percentage would be somewhat larger if the comparison were made on the number of investigations rather than shipments, it would still not be large compared to the total. Does this mean then that chemistry can derive little benefit from the use of isotopes? Indeed, not.

Instead, it means that chemists have been so preoccupied in assisting isotope investigations in other fields that they have not taken the opportunity to exploit the use of these newly available tools in their own problems. It also means that not enough chemists have obtained the proper training and experience to handle radioactive materials. Although training of this type has been available to chemists within the atomic project, it is not generally available outside the project. Until it does become widely available through the normal curricula of colleges and universities, the situation will undoubtedly continue to be a case of default by lack of numbers, not interest.

On the other side, it should be pointed out that chemistry is used in nearly every isotope tracer study undertaken. Indeed, isotope utilization is probably more dependent on chemistry than it is on physics.

CONTRIBUTIONS OF CHEMISTRY

The contributions of the chemist to the field of isotope utilization are many. They start with the basic contributions of nuclear chemistry research, follow through to the chemical aspects of isotope production, and finally make themselves more widely known in the applications and studies carried out with the materials.

Nuclear Chemistry Research. Nuclear chemistry has proved to be as necessary and fruitful as nuclear physics. For example, chemical identification is an extremely important and often necessary part of finding new isotopes. The first physicists to work with the cyclotron found that proper identification of the various isotopes produced during their nuclear studies often involved chemistry too complex for them to handle. Nuclear chemists had also been as active as physicists in measuring the radiation properties of a large number of radioisotopes. But perhaps most important, the chemist has developed the whole field of radiochemistry, that is, the chemistry of trace amounts of radioactive material. These contributions have, of course, been directly applicable to isotope utilization.

Radioisotope Production. Chemical processing may enter into three phases of radioisotope production; processing of target materials, processing of irradiated materials, and finally, processing of the radiomaterial as shipped in order that it may be used in a specific tracer study. The latter type of processing includes converting the radiomaterial into the proper chemical form or incorporating it into a complex labeled compound.

As with other phases of radiochemistry, the chemistry associated with the production and processing of radiomaterials is very exacting. Indeed, so exacting are many of the specifications that it is often necessary not only to purify the target materials to be irradiated but also the reagents used in the processing procedures. Although c.p. chemicals are used exclusively, sodium carbonate contains too much iron for certain separation procedures; nitric acid too much aluminum; hydrochloric acid too much lead; and aqueous ammonia too much silica.

In some instances, contaminants may be present in target materials in such low concentrations as not to be distinguishable by chemical means. For example, in the production of high-purity radionickel, all traces of cobalt must be removed from the stable nickel target. To date it has only been possible to guarantee that objectionable amounts of cobalt have been removed through a radiochemical analysis following irradiation of the target.

If the radiomaterial is formed by the neutron-gamma reaction the radioisotope formed is always the radioactive counterpart of the stable isotope used as the target material, and hence it is impossible to separate chemically the induced radioactive atoms from the residual stable atoms. In such instances, the choice and purity of the target materials are extremely important. Where feasible a pure form of the element is used. When a compound must be used, the target is selected so as to produce, in addition to the desired radioisotope, either (1) no other radioactive isotope, or (2) a short-lived radioisotope, or (3) a radioisotope which may be easily separated from the product isotope.

If a radioisotope is produced by transmutation or as a fission product, it must not only be separated from the irradiated material but from the other radioisotopes

present. A host of interesting problems have been encountered in the chemical processing of radioisotopes of this type, either because of the small amounts of radiomaterial handled or because the process has to be carried out remotely. In most cases less than a milligram of a radioisotope is extracted in a single batch from a very much larger amount of irradiated material. Often the amount of radioisotope is so small that it cannot be identified and measured except by the radiation which it emits. For example, one curie of short-lived radioiodine contains only 8×10^{-6} g. of the radioelement and even one curie of long-lived radiocarbon contains less than 250 mg. of the isotope. The number of radioactive atoms handled in some processes is so small compared to Avogadro's number that the material does not obey the mass action laws of stoichiometry.

In addition to the chemical purifications and separations that go into the production and processing of radioisotopes, the chemist is also responsible for converting the radiomaterial into a useful form. The many tracer uses made of radioisotopes have created demands for labeled compounds ranging from special modification of the simple basic compounds, through processing of the major intermediates, to the synthesis of complex compounds required for physiological and metabolic studies. To date, approximately 450 labeled compounds are known to have been synthesized by chemical means and more than 275 of them are routinely available.

Isotope Utilization. It is in the use of isotopic materials that the chemist has made the greatest number of contributions. Chemistry not only serves as the basis for a number of the unique techniques associated with the tracer method but also as the means by which tracer products are separated and identified. Indeed, the isotope tracer method depends very largely on chemistry.

Radioisotopes in their best known and most widely applicable role are used as tracer atoms. As such they serve as the basis for a new analytical technique—the tracer technique, which, because of its combination of sensitivity and unique specificity, is the most powerful analytical technique known today. The sensitivity of the method is so great that it is not difficult to work at dilutions as high as a billion (10^9) or ten billion (10^{10}) while dilutions of more than a trillion (10^{12}) are attainable. This would mean detecting a microgram uniformly mixed in over 1000 kg. or one ounce mixed in over 10 million tons!

In addition, the specificity of the technique permits the independent labeling and tracing of a specific batch of atoms or molecules even in the presence of other atoms or molecules or the same substance, and in spite of multiple reactions with numerous other kinds of atoms or molecules.

Another advantage of the tracer technique is that it is both qualitative and quantitative.

What is often overlooked, however, particularly by those who have not yet employed radioisotopes in their own research, is that the tracer technique does not always replace but often only supplements other analytical techniques. Also, if the tracer technique is to be used for quantitative analysis, one cannot usually eliminate the intermediate chemical steps associated with other analytical procedures. Because of the great sensitivity of radioisotope detection, the chemistry required in tracer analysis may often be even more exacting.

The chemist, working with the physicist, has developed three general ways in which radioisotopes may be used as analytical tools. These may be referred to as "tracer analysis," "isotope dilution analysis," and "activation analysis."

The first and simplest, tracer analysis, is designed to follow the fate of a radioelement or labeled material from one stage to a later stage of a reaction or process. It is primarily useful in determining the distribution of a specified material in a variety of end products. Depending on whether the radioactive label is added to the system in known amount, the technique may be either qualitative or quantitative. In either case, the advantages are that the determinations can be made at concentrations far below those permitted by other methods. If the technique is to be used quantitatively, the total amount of material being traced must be determined accurately in the final stage of the reaction process.

Isotope dilution analysis is a modification of tracer analysis. It is particularly suited for determining the amount of a substance in a process or system which is either present at a concentration too low to be measured by chemical or spectroscopic methods or which cannot be quantitatively separated from other materials in the system. The technique is based on introducing into the sample to be analyzed an amount of radioelement of known specific activity, that is, mixed in known ratio with stable atoms of the same element. Any change in this ratio will be due to the dilution caused by the amount of that element originally present in the system. The unknown amount of element originally present in the system is thus determined by the change in the ratio of radioactive to stable element. The advantage of the technique is that it does not require quantitative chemical separation and determination of the total amount of element in the system. It is only necessary to do quantitative work on a small sample, this sample, of course, being chemically pure.

Inverse isotope dilution is used when the labeled material is already present in the system. In this case a known amount of unlabeled compound is added and a sample subsequently removed for a specific activity measurement. The advantages here are that very small amounts or concentrations can be determined and the pure diluting compound added need not be labeled.

The double dilution technique is used when neither the amount or specific activity of the labeled material is known. By running two dilutions and solving two simultaneous equations, it is possible to eliminate one

of the unknowns and subsequently solve for both unknowns.

Activation analysis, which incidentally only recently became an integral part of the Commission's isotope distribution program and hence available for extensive use by non-Commission investigators, takes advantage of both nuclear physics and radiochemistry. It is based on the neutron irradiation of an unknown sample in the nuclear reactor, and the subsequent identification of the radioisotopes induced. The various radioisotopes thus produced are identified by their specific radiation characteristics. The technique can be either qualitative or quantitative and is particularly useful where the concentrations of the unknown element are too low to be identified by chemical or spectroscopic methods or where standard methods of analysis are not satisfactory because of interfering contaminants. Sensitivity of detection ranges from 10^{-6} to 10^{-11} g., that is, this is the amount of an element that will produce sufficient activity to be measured after the element has been exposed to a predetermined neutron flux for a given period of irradiation.

As has already been pointed out, the isotope tracer technique derives its power from its great sensitivity and specificity. Sensitivity is primarily concerned with the detection and measurement of radioactivity and hence is not the primary responsibility of the chemist. Specificity, on the other hand, depends on careful separation and identification procedures, which are chemical techniques. Further, the final radioactivity measurements in a tracer experiment are only as significant as the preciseness of the techniques used to separate and identify the samples.

The chemist has had to devise new methods and adapt others to meet these strict separation and identification prerequisites. Those employed to date include ion exchange, chromatographic, and isotope derivative techniques. Although time does not permit a detailed discussion of the ways in which these three techniques have been used, a brief description of the latter two may be in order since they may not be as widely known.

Paper chromatography has proved a particularly useful technique since it permits separation and identification to be made in the same operation. The unknown compounds are permitted to move in a solvent along a strip of filter paper. As in the case of column chromatography, individual compounds or mixtures of compounds separate out at various distances from the point at which the solvent was added. Identification of the separated compounds may then be made by comparison with known chromatograms or by a combination of radiochemical and microchemical techniques. Also, autoradiography has been found to be a helpful adjunct to paper chromatography. Recently, two-dimensional paper chromatography has been introduced into isotope work. Two-dimensional chromatography offers the advantage of a second order of separation. After one solvent has caused the products of an unknown mixture to move along one dimension of the paper, a second solvent is added in such a manner that the newly separated products move in a second path at a right angle to the path of the original separation.

The isotope derivative method, like paper chromatography, will probably find widest application in biological tracer studies. Briefly, it is a method for estimating organic compounds in the form of isotopically labeled derivatives. To a mixture containing the compounds to be analyzed is added a reagent containing an isotope under such conditions that the compounds in question are converted into isotopic derivatives of the reagent. The isotope derivative method becomes quantitative (1) when quantitative yields are obtained of the isotopic derivatives during the reaction with the isotopic reagent as well as in subsequent isolations or (2) when a known amount of the compound being analyzed for, labeled with a second isotope, is introduced into the mixture as an indicator. The recovery of the second isotope in any pure isolated sample of the isotopic derivative may be used to correct both the lack of quantitative yield and recovery obtained with the derivative of the original isotopic mixture.

For example, to determine glutamic acid in an amino acid mixture, one reacts the mixture with a certain radioiodine-labeled reagent, adds a known amount of very pure radiosulfur-labeled glutamic acid, and then separates the amino acids by chromatography. If the ratio of counts of radioiodine to radiosulfur is determined in successive small strips over the glutamic acid band, one can identify with certainty and measure the amount of glutamic acid in the original mixture.

And throughout the course of all tracer experiments the isotope user must be continually aware of pitfalls. Potential pitfalls lie in each of the seven criteria of tracer methodology, namely: radiochemical purity, single chemical state, exchange error, degree to which the tagged molecule remains intact, isotope effect, chemical effects, and radiation effects. It is primarily the responsibility of the chemist to see that such pitfalls don't invalidate the results of tracer experiments.

FUTURE

The future extent of isotope utilization will, of course, depend on a number of factors. For example, it will depend on the continued and expanded availability of isotopic materials, on more scientists acquiring the training to handle and use radioisotopes, and on a wider applicability of the tracer technique. Taking each of the factors separately, we find: first, that future radiosotope production will be sufficient to meet all demands, since nuclear reactors have proved their unique ability as production units and more reactors are continually being put into operation; second, even though lack of trained people is still a major bottleneck, this situation will gradually improve by informal training if not by formal training; and third, scientists in increasing numbers are gaining a general "feeling" for the tracer method to the extent that it is becoming common practice in attacking research problems to see

whether the isotope technique can be applied advantageously.

Perhaps the most important long range factor, however, is the question of whether the power of the tracer technique can be improved. For instance, have we arrived at the ultimate in the ability to trace atoms? Is there any possibility of pushing forward the limits of the technique?

It is only by pushing forward the working limits of sensitivity of measurement, and the specificity of separation and identification techniques that the tracer method can be made more productive.

Physical and Chemical Advances. The principal areas in which the tracer method can be improved by "physics" are production and instrumentation. There have been a number of recent achievements in both areas. For example, the development of high-flux reactors makes it possible to increase the specific activity of radiomaterials produced by neutron-gamma reactions. With an increase in the atom per cent of radioactivity in a tracer material, greater dilutions are permitted in its use.

The sensitivity of instruments now available permits radiation measurements that are only slightly above background. Using the readily obtainable sensitivity of present commercial instruments and without carrier-free materials, dilutions as high as 10^9 and 10^{10} have been possible. It has recently become possible by using anticoincidence methods, such as are used in carbon 14 dating and tritium water analysis studies, to count down to a fraction of the cosmic-ray background. Other recent improvements in instrumentation include the development of liquid scintillation counters and improved gas-filled counters.

The combined effect of these improvements in production and instrumentation means that from a purely physical aspect the tracer method is even now being pushed to its utmost—to the theoretical limits of dilution which as previously noted may be greater than a trillion. Certain biological studies have already begun to demand that measurements be made at the greatest possible dilutions. Although further refinements will undoubtedly be made, it is doubtful that these will be able to push forward significantly the physical working limits.

We have already noted the contributions of chemistry to the advancement of the tracer technique. Many more, however, remain to be made. A number of newer techniques have not as yet been completely developed or extensively exploited. Of special note are double isotopic labeling, two dimensional paper chromatography, the isotope derivative method and activation analysis. Since the working limits of the tracer method, as far as the physical aspects are concerned, are already being pushed to their utmost, most of the improvements to be made in the future will have to be in the form of chemical advances.

As the physical and chemical techniques associated with the tracer method are advanced, isotope utilization will expand because more and more problems will become subject to study by the tracer method. To make such advances, however, will require the efforts of more investigators. This leads us to the obvious conclusion that we will have to train more chemists in the field.

TRAINING

Numerous references have been made to the need for more chemists trained in handling and using radioisotopes. The radioisotope is one of the most valuable analytical tools now known, yet the chemist is seldom exposed to it during his educational training. The problem, however, is not as simple as it may first appear.

First, with a few exceptions, such training is not now available. Chemists working with the atomic energy project, of course, obtained experience in handling and using radioactivity. Outside Commission programs, a limited number obtain the necessary rudiments through the basic training course in isotope techniques offered by the Oak Ridge Institute of Nuclear Studies. An even smaller number are fortunate enough to obtain such training in one of the few universities and colleges now offering students the opportunity to study radiochemistry and isotope tracer techniques in formal graduate courses.

There is no simple formula for immediately filling this need. A lecture-laboratory course incorporated into the curriculum as part of the analytical course work now carried by the chemistry student would be a step in the right direction. It is difficult to say whether such training should be made part of the undergraduate or graduate work. Perhaps it should be offered at both levels. In either case, it would increase the number of chemists who would be able to determine wherein radiosotopes might be used to advantage in any future research in which they might participate.

But the training problem is much broader than this. The same training should also be made available to the students of physics, biology, and other sciences as well. Ideally, the course should be suited for students of different majors. Such a course might logically be offered by the chemistry department. Care should be taken to avoid having the general "isotopes techniques course" become largely a "radiochemistry course," thereby scaring away the physics and biology students. Perhaps this could be done through an interdepartmental effort with faculty representation for each of the departments concerned.

In any event, the isotope technique places a real responsibility on the educator.

CLOSING REMARKS

In our emphasis on the problems associated with isotope utilization and chemistry's essential role, we have intentionally touched only briefly on the outstanding accomplishments of isotopes. In closing, however, we should like to point out a few of these accomplishments.

Physiologists have been able to develop new concepts of the dynamic state of body constitutents and to de-

velop an entirely new technique for studying the synthesis, transport, utilization, and breakdown of various body components. Isotopes have contributed immensely to our understanding of photosynthesis and it is now apparent that an understanding of this basic conversion of solar energy to chemical and life energy may some day have great consequences. Isotopes have shed considerable light on our knowledge on the mechanics of chemical and physical-chemical reactions. In medicine, isotopes have not only proved a valuable tracer in clinical research but also have proved in a number of instances to be useful diagnostic and therapeutic aids. And finally, isotopes have helped solve problems that could be solved in no other way in nearly every field of scientific and technological development.

Foreseeing an abundant supply of suitable isotopic materials, foreseeing rapid physical and chemical advances to the ultimate limits of the isotope technique, and finally foreseeing educators meeting the need for more trained specialists, we can predict much more extensive and valuable use of radioisotopes in the years to come.

EDWARD DIVERS AND THE CHEMISTRY OF NITROGEN COMPOUNDS[1]

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EDWARD DIVERS was born in London, of Kentish ancestry, on November 27, 1837 (*1*). He was one of three children. As an infant he had an attack of inflammation of the eyes. This illness weakened his vision permanently, and the defect could not be remedied by eyeglasses. Years later an explosion in his Japanese laboratory made matters much worse, and Divers lost almost the entire sight of his right eye. Despite this great handicap, Divers left behind him an impressive record of detailed research investigations involving the keenest powers of observation.

At the age of 13, Divers entered the City of London School. One of the teachers there was Thomas Hall, a former student of A. W. Hofmann at the Royal College of Chemistry. Hall gave experimental lectures in chemistry between regular class hours, and attendance at these was voluntary. Divers was an interested student at these lectures.

From the School Divers moved on to the Royal College of Chemistry, then located in Oxford Street, and studied for one year under Hofmann. A fellow-student was William Perkin, and an assistant on the staff was William Crookes, from whom Divers received valuable training in exact methods of investigation.

Divers became a junior assistant in Stenhouse's laboratory in St. Bartholomew's Hospital Medical School the following year. Here again he worked among future scientists of note, for Abel was then an assistant, and both Kekulé and H. Buff worked at the same benches with Divers.

It has been pointed out elsewhere that there was a close link between the Royal College of Chemistry and the various Queen's Colleges which Prince Albert was responsible for founding in various Irish centers at mid-century (*2*). The early holders of the chemistry chairs in these new colleges were largely members of Hofmann's original group. Edmund Ronalds (*3*), who had been assistant to Liebig before working with Hofmann, became professor of chemistry in Queen's College, Galway, in 1849. Five years later he asked Divers to join him as an assistant. Thomas Rowney, another of the Hofmann school, succeeded Ronalds in 1856, and Divers continued as his assistant also.

Divers accepted the Irish appointment with two objectives in mind. First, he wanted to take the University degree in medicine, one of the few scientific degrees then available. Secondly, he wanted to take advantage of the opportunities offered for teaching and for research in chemistry. He achieved both ends, for he was awarded his M.D. degree in 1860 and he spent some 12 years in Galway as assistant and demonstrator in chemistry. He had a natural ability as a teacher and had a reputation as a popular lecturer, giving series of public lectures in chemistry for the first time in various towns in the west of Ireland.

He left Ireland permanently in 1866 and held a number of teaching appointments in England in the years

[1] Presented before the Division of the History of Chemistry at the 122nd Meeting of the American Chemical Society, Atlantic City, New Jersey, September, 1952.

immediately following. He was at various times a lecturer in materia medica at Queen's College, Birmingham, in physics at Charing Cross Hospital Medical School, and in medical jurisprudence at Middlesex Hospital Medical School. This latter post gave him a background in forensic medicine which made him something of an expert on scientific criminology in later years. He was also lecturer in chemistry at the Albert Veterinary College and contributed special reports on the quality of the London milk supplies.

In 1873, at the invitation of the Public Works Department of Japan, he and ten other British specialists went to that country to found a College of Engineering. Japan had just begun the task of remodeling its civilization by the systematic introduction of western sciences and institutions, and as part of the plan a series of schools of liberal and technical education were planned for the Tokyo area.

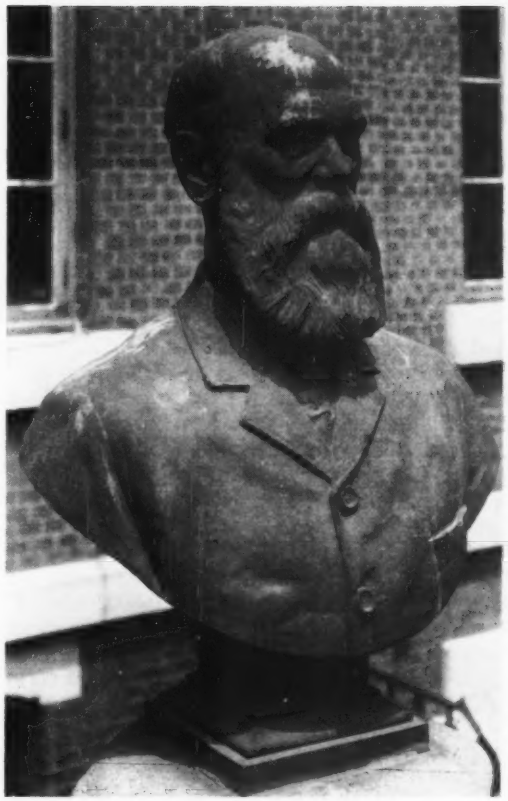
Divers' task was the teaching of general and applied chemistry. His original appointment was for five years, but in all he spent 26 unbroken years in Japan. In 1882 he became principal of the College as well as professor of chemistry. In 1886 the Engineering College was merged into Tokyo University, where Divers was professor of inorganic chemistry till 1899 when he returned to England.

The first seven or eight years of his Japanese appointment were fully occupied with supervising the building of his laboratories, a heavy teaching schedule, and the execution of a large variety of chemical and mineralogical analyses for various government departments. However, from 1881 onward the volume of his research papers reached substantial proportions. In addition he trained and inspired with his own enthusiasm a school of younger chemists who won notable recognition later. These included such well-known names as Haga, Takamine, Shimidzu, and Nakamura. His colleague Sakurai wrote from Japan after Divers' death that he "will be forever remembered in this country as the great promoter of chemical science"(*4*).

Two personal tragedies marked Divers' home life in Japan. First his only son, Frederic, who was a member of the Chinese customs service, died of a sudden illness in Amoy. Then his wife, the former Margaret Theresa Fitzgerald of Mayfield, Cork, Ireland, died in Tokyo in 1897. He had two daughters, but both of these were married in Japan, one to a French military attaché and the other to an English businessman. Lonely and in failing health, he decided to return home in 1899.

On leaving Japan he was made professor emeritus of the University of Japan. In 1886 the Japanese government had honored him with the order of the Rising Sun of the Third Class. To this was added on his retirement the Order of Sacred Treasure of the Second Class. At the entrance to the College of Science in Tokyo his many friends erected a bronze memorial likeness in his honor (*5*).

Back in England Divers took an honored place in chemical affairs. His house was described as the Mecca of Japanese students visiting London. In 1885 the



Bronze Likeness of Edward Divers at Entrance to College of Science in Tokyo

Royal Society had elected him a Fellow in recognition of his important contributions to the general progress of chemistry. His old University in Ireland, represented by the Royal University, awarded him the doctor of science degree *honoris causa.* He was vice-president of the Chemical Society in 1900; president of the chemical section of the British Association in 1902; president of the Society of Chemical Industry and vice-president of the Institute of Chemistry in 1905 (*6*).

In his later years Divers was remembered best as the central figure of groups of younger workers who looked to him as a leader, particularly in the fields he had made his own. A few years before his death he revisited Galway and, although almost blind, he took the greatest interest in feeling his way through the laboratories where he had worked half a century earlier. He discovered in the basement the remains of the old apparatus and equipment which he had once used in his classes (*7*).

Following an operation and a painful illness Edward Divers died at his home in Kensington on April 8, 1912, and was interred at Brookwood, Surrey. Notable among the many tributes paid to him by his former students and associates was that of Sakurai, who wrote from Japan: "Divers spent the best part of his

life in this country and by his teaching and example during this long and meritorious service he greatly promoted the development of chemical science in Japan, besides contributing very largely to the general progress of chemistry" (*8*).

SCIENTIFIC WORK

The year in which Divers received the M.D. degree in Queen's College, Galway, he joined the Chemical Society. Two years later he published his first paper, a careful study of magnesium ammonium carbonate; and six years later, in 1868, he published a similar study of zinc ammonium chloride. In 1870 he published three papers describing his classical investigation of the carbonates and carbamate of ammonium. He showed for the first time (*9*) that there were at least three well-defined ammonium carbonates, the normal carbonate, the hydrocarbonate, and the sesquicarbonate. His study of preparative methods, composition, and reactions of ammonium carbamate (*10*) was characterized by extreme accuracy and exceptional keenness of observation.

In 1863 Divers had studied the spontaneous change which guncotton undergoes with the formation of gelatinous acids, and then in 1871 appeared two papers in which he began the study of nitrites and announced his discovery of the hyponitrites. He found (*11*) that when a solution of sodium nitrite or nitrate is treated with sodium amalgam the nitrite or nitrate is reduced to sodium hyponitrite:

$$2NaNO_3 + (8H) = 4H_2O + Na_2N_2O_2$$

In 1873 he studied the interaction of ammonia and ammonium nitrate. These studies were later to be extended very considerably during his time in Japan.

As pointed out earlier Divers' first years in Japan were fully occupied in placing his department on a sound footing. However, from 1883 onward his name appeared with great frequency in the scientific journals.

His first original research work in Japan was in connection with tellurium and selenium ores which he found associated with Japanese sulfur deposits, and which he later obtained in some quantity from the lead chamber deposits of the Osaka sulfuric acid plant (*12*). The difficult problem of separating small amounts of selenium from impure tellurium ores had occupied the attention of Berzelius and many other chemists. Rose (*13*) had observed that selenium is imperfectly precipitated in the presence of sulfuric acid alone, but Divers and his assistant Shimosé showed that this was true only in dilute acid, for precipitation is complete when sufficiently concentrated acid is used. Based on this observation they devised a new and simple method for the separation of selenium and tellurium (*14*). They also investigated tellurium monoxide and showed for the first time that when heated in hydrochloric acid gas it is converted to tellurium dichloride. They also studied extensively tellurium sulfotrioxide ($TeSO_3$), which is a product of the reaction of sulfur trioxide and tellurium (*15*).

Divers' activity as an investigator became suddenly prominent about 1884–85 when he published no less than 24 papers in the *Journal of the Chemical Society* alone. The majority of these, and of his subsequent papers, deal with the chemistry of nitrogen and sulfur compounds, a subject to which he contributed more than any other single chemist. In the compass of this study it is impossible to summarize even briefly the immense amount of work done by Divers in the field, and little more can be attempted beyond a listing of the more significant highlights.

In a continuation of his earlier work on hyponitrites Divers, in collaboration with Haga (*16*), established the composition of silver hyponitrite to be $(AgNO)_2$ against the formula $Ag_4N_4O_5$ asserted to be true by Berthelot and Ogier. Several new modes of formation of hyponitrites were discovered at the same time. Interesting among these was the ready decomposition of an oxyamidosulfonate (which we would nowadays designate as a salt of hydroxynitrilodisulfonic acid) by excess concentrated alkali to yield a hyponitrite and a sulfite:

$$2N(OK)(KSO_3)_2 + 4KOH = (KON)_2 + 2K_2SO_3 + 2H_2O + 2K_2S0$$

This reaction afforded proof that in hyponitrites the metallic radical is not in direct union with nitrogen, but is linked to it through oxygen. The reaction is also significant in that it gives a means of preparing large quantities of hyponitrites, and is in fact the basis for Kirschner's method of preparation.

In another paper (*17*) Divers and Haga made a strong case for the sulfonic or unsymmetrical constitution of inorganic sulfites. One of his main points was the experimental fact he established that, when phosphorus oxychloride acts on sodium sulfite, one half of the sodium goes to sodium chloride while the other half goes to sodium phosphate:

$$3SO_2(Na)(ONa) + POCl_3 = 3NaCl + (NaO)_3PO + 3SO_2$$

The only experimental evidence in favor of a symmetrical constitution for the sulfites available at that time lay in the deduction of Carius that the thionyl chloride formed by the action of phosphorus pentachloride on inorganic sulfite was a direct product of the reaction. Divers pointed out that this view was incorrect in that the thionyl chloride was evidently merely the result of a secondary reaction between sulfur dioxide and phosphorus pentachloride.

More recent work (*18*) seems to indicate that there is as yet no definite evidence of the existence of isomeric potassium sodium sulfites, namely, $KO.SO_2.Na$ and $NaO.SO_2.K$. The inference, therefore, is that the graphic formulas of the sulfites correspond to two symmetrically placed hydroxyl groups in sulfurous acid representing it graphically as HO—SO—OH. In addition, Pascal's work (*19*) on magnetic observations favors the $SO(OH)_2$ formula. However, it is worth noting that the negative results obtained to date in attempts to prepare isomeric potassium sodium sulfites do not prove conclusively that an unsymmetrical

acid does not exist, but may merely indicate that there is only one stable or favored configuration of the salt under the specific experimental conditions.

It was in the course of this work that Divers suffered the eye injury mentioned earlier. On November 24, 1884, a bottle in which phosphorus oxychloride was kept in the laboratory exploded violently, and he was badly cut about the face by glass splinters. For a time it was feared that he would lose his sight altogether, but he eventually regained partial vision. Though he could no longer recognize friends when he met them in the street, he could read with one eye at close range. Despite this serious handicap the pace of his work accelerated rather than diminished. In fact, the accident which cut him off from so many normal interests tended to make him concentrate even more on scientific work. In vacations as well as during the school year he spent almost all his waking hours at his bench in the laboratory.

In one aspect of his work on the sulfites he collaborated with Shimidzu on an exhaustive study of the numerous sulfites of mercury, a work which still stands as the major report in his highly complex field (*20*). Divers and Shimidzu also investigated the calcium hydrosulfites (*21*) and obtained solid calcium hydrosulfide for the first time. This paper showed, *inter alia*, the constitution of the compound formed (Ca(OH)(SH)) from slaked lime in sulfide purifiers.

Fulminic acid and hydroxylamine also attracted a good deal of Divers' attention. He and Kawakita showed (*22*) that formic acid formed a significant part of the products of decomposition of mercury fulminate by hydrochloric acid. This fact had been overlooked by earlier workers who were puzzled by the fate of the carbon of the fulminates on decomposition. Divers showed in a study of the formation of hydroxylamine from nitric acid (*23*) that of all the common metals zinc or tin is best suited for effecting the reduction.

Divers and Shimidzu (*24*) believed that, in the reduction of dilute nitric acid by zinc, ammonia and hydroxylamine are not consecutive products but are formed by parallel and independent reactions. They maintained that hydroxylamine when once formed is not readily reduced to ammonia, and is therefore not an intermediate stage in the reduction to ammonia. They also showed that the presence of sulfuric acid favored the formation of hydroxylamine, but no explanation of this peculiar effect has yet been advanced. By using a considerable excess of sulfuric acid they found, in accord with Kinnear's observations (*25*), that nearly all, if not all, the nitric acid not converted to hydroxylamine is converted to ammonia.

Probably Divers' most intensive series of studies were those carried out on the chemistry of sulfonated nitrogen compounds. With Haga (*26*) he showed that the numerous complex acids belonging to this group of compounds were the products of the reaction between sulfurous and nitrous acids, the base being essential only in so far as it protects the products of the reaction against hydrolysis, and that, contrary to the views of earlier workers, normal sulfites and nitrites have no action whatever on each other. They also showed that the primary product of the interaction of sulfurous and nitrous acids is always hydroxylaminedisulfonic acid and nothing else, no derivative of dihydroxylamine or of any quinquevalent nitrogen ever being formed. All the substances supposed by others to be such were proved to be merely double salts of hydroxylaminedisulfonate and alkali nitrite, with or without alkali replacing the hydroxyl hydrogen of the disulfonate, and to be obtainable by synthesis from hydroxylaminedisulfonate, alkali nitrite, and alkali hydroxide (*27*). The chemistry of these rather complex compounds was thus considerably simplified.

In his later years in England the record of Divers' work on the oxides of nitrogen and sulfur made him a recognized authority on the reactions in sulfuric acid chambers and on the composition of alkali waste (*28*).

The record of the chemical work carried out by Edward Divers would in itself be noteworthy. The part which he played in shaping the destinies first of the new chemical school of Queen's College, Galway, Ireland, and later of the University in Tokyo, Japan, make him of even greater significance in the development of modern chemical education. In an era that produced many of the great men of chemistry, the name of Edward Divers deserves also to be remembered.

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THE ALGEBRA OF SIMULTANEOUS EQUILIBRIA

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A MAJOR part of the theoretical background for qualitative analysis consists of the six fundamental types of equilibria which occur in aqueous solutions of electrolytes. Three of these types of equilibria involve acid-base reactions; two solubilities; and one, oxidation reduction. These reversible processes and their equilibrium constant expressions are exemplified in Table 1.

TABLE 1

Equilibrium constant	*Reversible process*	*Equilibrium constant expression*
K_a	Acidity constant (typified): $HA + H_2O \rightleftarrows H_3O^+ + A^-$	$K_a = \frac{[H_3O^+][A^-]}{[HA]}$
K_b	Basicity constant (typified): $H_2O + A^- \rightleftarrows HA + OH^-$	$K_b = \frac{[HA][OH^-]}{[A^-]}$
K_i	Instability constant; *e. g.*, $Ag(NH_3)_2^+ \rightleftarrows Ag^+ + 2NH_3$	$K_i = \frac{[Ag^+][NH_3]^2}{[Ag(NH_3)_2^+]}$
K_{sp}	Solubility product; *e. g.*, (solid)$SrSO_4 \rightleftarrows Sr^{++} + SO_4^{--}$	$K_{sp} = [Sr^{++}][SO_4^{--}]$
K_s	Intrinsic solubility of a weak electrolyte; *e. g.*, (gas; 1 atm.)$H_2S \rightleftarrows H_2S$ (dissolved)	$K_s = [H_2S]$
K_r	Oxidation-reduction; *e. g.*, $Fe^{3+} + Fe(CN)_6^{4-} \rightleftarrows Fe^{++} + Fe(CN)_6^{3-}$	$K_r = \frac{[Fe^{++}][Fe(CN)_6^{3-}]}{[Fe^{3+}][Fe(CN)_6^{4-}]}$

Numerous problems arise in which *two* of these fundamental types of equilibria or two different examples of the same fundamental type are involved simultaneously. For example, the solubility of silver chloride in ammonia solutions depends upon both the *solubility product* of silver chloride and the *instability constant* of diamminesilver ion. In such cases, at least one dissolved species is common to the two reversible processes involved (in the example given, silver ion is the common species). The two equilibria may, therefore, be referred to as *simultaneous equilibria.*

The conventional method of solving such problems has recently been criticized by Boyd[1] because it entails solving one of the equilibrium constant expressions to obtain the value for the concentration of the common species and then substituting this value in the second equilibrium constant expression. The student, complains Boyd, "is expected to see that two equilibria are involved." In addition Boyd objects to the unnecessary work of calculating the concentration of the common factor. (This may, of course, be avoided by solving the first equilibrium constant expression algebraically and then substituting into the second expression before introducing numerical values.)

Boyd has developed a procedure in which he writes the overall equation and a corresponding overall equilibrium constant expression. In order to evaluate this constant, however, he must multiply the expression algebraically by the ratio, *common species/common species,* or some power of it. This species is not selected for the reason that it is recognized to be the common species in two equilibria but simply because it is "obvious" that the introduction of this species will allow a known equilibrium constant expression to be separated from the resulting algebraic expression, leaving a residual expression, which is then recognized as some other equilibrium constant expression (or a power or reciprocal thereof).

[1] BOYD, R. N., J. CHEM. EDUC., **29**, 198 (1952).

Both Boyd's method and the conventional one suffer from the drawback of operating on equilibrium constant expressions. The present paper proposes a method which employs the simpler process of operating on the *chemical equations* themselves together with the *symbols* for the equilibrium constants (not their expressions). The possible value of this approach is illustrated below in a systematic survey of cases of simultaneous equilibria and by some typical problems.

RULES FOR SIMULTANEOUS EQUILIBRIA

The proposed method for dealing with simultaneous equilibria involves a simple algebra which is described by the following rules.

(1) *If a chemical equation is written in reverse the equilibrium constant for this reaction is the reciprocal of the equilibrium constant as usually written.* Thus if

$$HA + H_2O \rightleftarrows H_3O^+ + A^-$$

has the constant, K_a, then

$$H_3O^+ + A^- \rightleftarrows HA + H_2O$$

has the constant, $1/K_a$.

(2) *If the coefficients in a chemical equation are multiplied by an integer,* n, *the equilibrium constant for this process is the nth power of the usual equilibrium constant.* Thus, referring to the example given in Rule 1,

$$2HA + 2H_2O \rightleftarrows 2H_3O^+ + 2A^-$$

has the constant, K_a^2.

(3) *If the chemical equations for two simultaneous re-*

actions are added, the equilibrium constant for the resultant reaction is the product of the equilibrium constants for the separate reactions. The equations should be written in such a way that the common species appears on opposite sides in the two equations and with the same coefficient (Rules 1 and 2 are applied). The overall equilibrium constant *expression* may be obtained from the overall equation in the usual way. The simple operation outlined in Rule 3 is the equivalent of solving one of the two equilibrium constant expressions algebraically for the common species and then substituting the result in the second equilibrium constant expression, as may be done in the conventional method. In such systems, *three* equilibrium constants are satisfied, the two simple ones and the overall constant.

Recognition of the principles of simultaneous equilibria allows a systematic survey of cases. These may involve not only two different fundamental types of equilibria but also two examples of the same fundamental type. More than two simultaneous equilibria may be added in one operation by a simple extension of Rule 3. The mathematically possible cases of simultaneous equilibria involving the six fundamental types taken two at a time are given in Table 2. One of these (K_s-K_s) is not chemically possible.

TABLE 2

K_a-K_a					
K_a-K_b	K_b-K_b				
K_a-K_i	K_b-K_i	K_i-K_i			
K_a-K_s	K_b-K_s	K_i-K_s	K_s-K_s		
K_a-K_{sp}	K_b-K_{sp}	K_i-K_{sp}	K_s-K_{sp}	K_{sp}-K_{sp}	
K_a-K_r	K_b-K_r	K_i-K_r	K_s-K_r	K_{sp}-K_r	K_r-K_r

ACIDS AND BASES

K_a-K_a. Diprotic acids of the type, H_2D, undergo successive protolyses. These constitute simultaneous equilibria. Thus,

Process	*Constant*
$H_2D + H_2O \rightleftarrows H_3O^+ + HD^-$	K_{a_1}
$HD^- + H_2O \rightleftarrows H_3O^+ + D^{--}$	K_{a_2}
Adding, $H_2D + 2H_2O \rightleftarrows 2H_3O^+ + D^{--}$	$K_{a_1}K_{a_2}$

that is,

$$K = \frac{[H_3O^+]^2\,[D^{--}]}{[H_2D]} = K_{a_1}K_{a_2}$$

The overall protolysis, therefore, has a constant which is equal to the product of the first and second protolysis constants. Furthermore, if $[H_2D] = [D^{--}]$ (which is true in an aqueous solution of NaHD), then

$$[H_3O^+]^2 = K_{a_1}K_{a_2}$$

and

$$[H_3O^+] = \sqrt{K_{a_1}K_{a_2}}$$

or

$$pH = {}^1/_2\,(pK_{a_1} + pK_{a_2})$$

By reversing the second equation another important relationship is obtained.

Process	*Constant*
$H_2D + H_2O \rightleftarrows H_3O^+ + HD^-$	K_{a_1}
$H_3O^+ + D^{--} \rightleftarrows H_2O + HD^-$	$1/K_{a_2}$
$H_2D + D^{--} \rightleftarrows 2HD^-$	K_{a_1}/K_{a_2}

that is,

$$K = \frac{[HD^-]^2}{[H_2D]\,[D^{--}]} = \frac{K_{a_1}}{K_{a_2}}$$

Hence, in a solution of NaHD in which $[H_2D] = [D^{--}]$

$$\frac{[HD^-]}{[H_2D]} = \sqrt{\frac{K_{a_1}}{K_{a_2}}}$$

The ratio, $[HD^-]/[H_2D]$, is a measure of the extent to which HD^- is converted to a mixture of H_2D and D^{--}.

These relationships are useful in calculating the pH's of solutions of "acid salts" such as $NaHCO_3$ and $NaHSO_3$ as well as the isoelectric points of ampholytes of zero net charge such as glycine, $^+NH_3CH_2COO^-$. They may also be employed to obtain the proportion of diprotic acid and base in equilibrium with the "acid salt" or ampholyte.

Problem 1. Calculate the pH of a solution of sodium bicarbonate and the ratio of carbonate ion to bicarbonate ion.

Solution:

$$pH = {}^1/_2\,(pK_{a_1} + pK_{a_2}) = {}^1/_2\,(6.37 + 10.33) = 8.35$$

The ratio,

$$[CO_3^{--}]/[HCO_3^-] = \sqrt{K_{a_2}/K_{a_1}} = \sqrt{10^{-10.33}/10^{-6.37}} = 10^{-1.98} = {}^1/_{100}\ \text{(approx.)}$$

Similar relationships are obtained for the reaction of an acid, HA, with a base, B, for which the simultaneous equilibria are shown below.

Process	*Constant*
$HA + H_2O \rightleftarrows H_3O^+ + A^-$	K_{a_1}
$H_3O^+ + B \rightleftarrows BH^+ + H_2O$	$1/K_{a_2}$
$HA + B \rightleftarrows BH^+ + A^-$	K_{a_1}/K_{a_2}

If $[HA] = [B]$, then $[BH^+] = [A^-]$ and

$$\frac{[BH^+]}{[HA]} = \sqrt{\frac{K_{a_1}}{K_{a_2}}}$$

The pH at equilibrium when equivalents of HA and B are reacted may be obtained by reversing the second equation of the two given just above.

Process	*Constant*
$HA + H_2O \rightleftarrows H_3O^+ + A^-$	K_{a_1}
$BH^+ + H_2O \rightleftarrows H_3O^+ + B$	K_{a_2}
$HA + BH^+ + 2H_2O \rightleftarrows 2H_3O^+ + A^- + B$	$K_{a_1}K_a$

If $[HA] = [B]$ and $[BH^+] = [A^-]$, then

$$[H_3O^+]^2 = K_{a_1}K_{a_2}$$

and

$$[H_3O^+] = \sqrt{K_{a_1}K_{a_2}}$$

or

$$pH = {}^1/_2\,(pK_{a_1} + pK_{a_2})$$

that is, the pH at equilibrium in reactions between equivalent quantities of HA and B is equal to one-half the sum of the two pK_a's (*i. e.*, those of HA and BH^+).

Problem 2. One equivalent of ammonia is mixed with one equivalent of hydrocyanic acid in dilute solution. Calculate the pH of the solution and the ratio of ammonium ion to hydrocyanic acid.

Solution:

$$pH = {}^1/_2\,(pK_{a_1} + pK_{a_2}) = {}^1/_2\,(9.4 + 9.25) = 9.33$$

The ratio,

$$[NH_4^+]/[HCN] = \sqrt{10^{-9.4}/10^{-9.25}} = 1 \text{ (approx.)}$$

K_a-K_b. The equilibria involved in the acidity and basicity constants of an acid, HA, and of its conjugate base, A^-, are simultaneous equilibria.

Process	*Constant*
$HA + H_2O \rightleftarrows H_3O^+ + A^-$	K_a
$A^- + H_2O \rightleftarrows HA + OH^-$	K_b
$H_2O + H_2O \rightleftarrows H_3O^+ + OH^-$	K_aK_b

hence,

$$K = [H_3O^+]\,[OH^-] = K_w = K_aK_b = 10^{-14}\ (25°)$$

and

$$pK_a + pK_b = 14\ (25°)$$

K_b-K_b. The over-all basicity constant of a diprotic base, D^{--}, is equal to the product of the first and second basicity constants.

Process	*Constant*
$D^{--} + H_2O \rightleftarrows HD^- + OH^-$	K_{b_2}
$HD^- + H_2O \rightleftarrows H_2D + OH^-$	K_{b_1}
$D^{--} + 2H_2O \rightleftarrows H_2D + 2OH^-$	$K_{b_2}K_{b_1}$

The stability of the ampholyte HD^- may be expressed in terms of K_b's. Thus, by reversing the second equation the following result is obtained.

Process	*Constant*
$H_2D + D^{--} \rightleftarrows 2HD^-$	K_{b_2}/K_{b_1}

(Compare the expression in terms of K_a's obtained above.)

K_i-K_i. The competition between two bases for a cation is illustrated by the reaction of cyanide ion on diamminesilver ion.

Process	*Constant*
$Ag(NH_3)_2^+ \rightleftarrows Ag^+ + 2NH_3$	K_{i_1}
$Ag^+ + 2CN^- \rightleftarrows Ag(CN)_2^-$	$1/K_{i_2}$
$Ag(NH_3)_2^+ + 2CN^- \rightleftarrows Ag(CN)_2^- + 2NH_3$	K_{i_1}/K_{i_2}

$$K = \frac{[Ag(CN)_2^-]\,[NH_3]^2}{[Ag(NH_3)_2^+]\,[CN^-]^2} = \frac{K_{i_1}}{K_{i_2}}$$

Problem 3. To a solution containing one equivalent each of cyanide ion and of ammonia is added 0.01 equivalent of silver ion. Calculate the ratio of the complex ions present in solution.

Solution:

$$[Ag(CN)_2^-]/[Ag(NH_3)_2^+] = 10^{-7.2}/10^{-22.1} = 10^{14.9}/1.$$

SOLUBILITIES

K_a-K_s. The overall equilibrium between a slightly soluble, monoprotic acid and its ions in solution is derived as follows.

Process	*Constant*
(solid) $HA \rightleftarrows HA$ (dissolved)	K_s
$HA + H_2O \rightleftarrows H_3O^+ + A^-$	K_a
(solid) $HA + H_2O \rightleftarrows H_3O^+ + A^-$	K_sK_a

hence,

$$K = [H_3O^+]\,[A^-] = K_sK_a$$

The overall constant ($= K_sK_a$) is the "solubility product" of weak or intermediate[2] monoprotic acids.

Problem 4. A slightly soluble, monoprotic acid has a solubility in water at 25° of 0.05 *M* and a K_a of 2×10^{-4}. Calculate its "solubility product."

Solution:

$$K_sK_a = 0.05 \times 2 \times 10^{-4} = 1 \times 10^{-5}$$

The solvent action of an acidic buffer on a slightly soluble, weak or intermediate base involves the following equilibria.

Process	*Constant*
(solid) $B \rightleftarrows B$ (dissolved)	K_s
$B + H_3O^+ \rightleftarrows BH^+ + H_2O$	$1/K_a$
(solid) $B + H_3O^+ \rightleftarrows BH^+ + H_2O$	K_s/K_a

hence,

$$K = \frac{[BH^+]}{[H_3O^+]} = \frac{K_s}{K_a}$$

and

$$[BH^+] = \frac{K_s[H_3O^+]}{K_a}$$

The "solubility" of the base is measured by the sum,[3]

$$[B] + [BH^+] = K_s + \frac{K_s[H_3O^+]}{K_a} = K_s\left(1 + \frac{[H_3O^+]}{K_a}\right)$$

Problem 5. A solid, monoprotic base has a K_b of 1×10^{-10} and a solubility in water of 0.001 *M*. Calculate its solubility in a buffer of pH 2.

Solution:

$$K_s\left(1 + \frac{[H_3O^+]}{K_a}\right) = 10^{-3}\left(1 + \frac{10^{-2}}{10^{-4}}\right) = 0.101\ M$$

(NOTE: $[BH^+] = 100 \times [B]$, so that the solubility is practically equal to $[BH^+]$.)

K_b-K_s. The "solubility product" of a slightly soluble, weak or intermediate base is derived as follows.

Process	*Constant*
(solid) $B \rightleftarrows B$ (dissolved)	K_s
$B + H_2O \rightleftarrows BH^+ + OH^-$	K_b
(solid) $B + H_2O \rightleftarrows BH^+ + OH^-$	K_sK_b

hence

$$K = [BH^+]\,[OH^-] = K_sK_b$$

The dissolution of a slightly soluble, weak or intermediate acid by a basic buffer involves the following equilibria.

Process	*Constant*
(solid) $HA \rightleftarrows HA$ (dissolved)	K_s
$HA + OH^- \rightleftarrows H_2O + A^-$	$1/K_b$
(solid) $HA + OH^- \rightleftarrows H_2O + A^-$	K_s/K_b

[2] DAVIDSON, D., *ibid.*, **19**, 154 (1942).
[3] DAVIDSON, D., *ibid.*, p. 221.

hence

$$K = \frac{[A^-]}{[OH^-]} = \frac{K_a}{K_b}$$

and

$$[A^-] = \frac{K_a[OH^-]}{K_b}$$

The observed solubility is measured by the sum,

$$[HA] + [A^-] = K_a + \frac{K_a[OH^-]}{K_b} = K_a\left(1 + \frac{[OH^-]}{K_b}\right) = K_a\left(1 + \frac{K_a}{[H_3O^+]}\right)$$

Problem 6. A solid, weak acid has a solubility in a buffer at pH 2 of 0.0001 *M* and in a buffer at pH 10 of 0.001 *M*. Calculate its K_a.

Solution:

$$0.001 = 0.0001\left(1 + \frac{K_a}{10^{-10}}\right)$$

$$K_a = 9 \times 10^{-10}$$

K_i-K_s. The solubility of a slightly soluble base, RNH_2, in an aqueous solution of a silver salt, for example, is governed by the following equilibria.

Process	*Constant*
(solid) $2RNH_2 \rightleftarrows 2RNH_2$ (dissolved)	K_s^2
$Ag^+ + 2RNH_2 \rightleftarrows Ag(RNH_2)_2^+$	$1/K_i$
(solid) $2RNH_2 + Ag^+ \rightleftarrows Ag(RNH_2)^+$	K_s^2/K_i

hence,

$$K = \frac{[Ag(RNH_2)_2^+]}{[Ag^+]} = \frac{K_s^2}{K_i}$$

K_b-K_{sp}. If the anion of a slightly soluble salt is a weak or intermediate base, the solubility product equilibrium is coupled with an acid-base equilibrium. For example, in the case of the solubility of barium carbonate the following equilibria are important.

Process	*Constant*
(solid) $BaCO_3 \rightleftarrows Ba^{++} + CO_3^{--}$	K_{sp}
$CO_3^{--} + H_2O \rightleftarrows HCO_3^- + OH^-$	K_{b2}
(solid) $BaCO_3 + H_2O \rightleftarrows Ba^{++} + HCO_3^- + OH^-$	$K_{sp}K_{b2}$

hence,

$$K = [Ba^{++}][HCO_3^-][OH^-] = K_{sp}K_{b2}$$

Problem 7. What is the pH of a saturated solution of barium carbonate in 0.02 *M* barium chloride?

Solution: Since HCO_3^- and OH^- ions are formed in equivalent amounts (see second equation above),

$$[OH^-]^2 = \frac{K_{sp}K_{b_2}}{[Ba^{++}]} = \frac{1.9 \times 10^{-8} \times 2.1 \times 10^{-4}}{2 \times 10^{-2}}$$

$$[OH^-] = 10^{-4.85}$$

$$pH = 9.15$$

The solubility of a slightly soluble hydroxide such as magnesium hydroxide in a solution of ammonium ion is governed by the following equilibria.

Process	*Constant*
(solid) $Mg(OH)_2 \rightleftarrows Mg^{++} + 2OH^-$	K_{sp}
$2NH_4^+ + 2OH^- \rightleftarrows 2H_2O + 2NH_3$	$1/K_b^2$
(solid) $Mg(OH)_2 + 2NH_4^+ \rightleftarrows Mg^{++} + 2H_2O + 2NH_3$	K_{sp}/K_b^2

that is,

$$K = \frac{[Mg^{++}][NH_3]^2}{[NH_4^+]^2} = \frac{K_{sp}}{K_b^2}$$

Problem 8. What ratio of ammonium ion to ammonia will prevent the precipitation of 0.1 *M* magnesium ion by ammonia?

Solution:

$$\frac{[NH_4^+]}{[NH_3]} = \sqrt{[Mg^{++}]\frac{K_b^2}{K_{sp}}} = \sqrt{\frac{0.1 \times 3.24 \times 10^{-10}}{1.2 \times 10^{-11}}} = 1.6$$

K_i-K_{sp}. The solubility of a slightly soluble salt such as silver chloride in a solution of ammonia is governed by the following equilibria.

Process	*Constant*
(solid) $AgCl \rightleftarrows Ag^+ + Cl^-$	K_{sp}
$Ag^+ + 2NH_3 \rightleftarrows Ag(NH_3)_2^+$	$1/K_i$
(solid) $AgCl + 2NH_3 \rightleftarrows Ag(NH_3)_2^+ + Cl^-$	K_{sp}/K_i

or

$$K = \frac{[Ag(NH_3)_2^+][Cl^-]}{[NH_3]^2} = \frac{K_{sp}}{K_i}$$

Problem 9. What concentration of ammonia is required to dissolve 0.01 mol. silver chloride per liter?

Solution:

$$[NH_3] = \sqrt{\frac{K_i[Ag(NH_3)_2^+][Cl^-]}{K_{sp}}} = \sqrt{\frac{6.8 \times 10^{-8} \times (0.01)^2}{1.6 \times 10^{-10}}} = 0.22\ M$$

Since this is the ammonia concentration at equilibrium and conversion to the complex is almost quantitative, that required initially is $0.22 + 2(0.01) = 0.24$ *M*.

Problem 10. Silver ion reacts with cyanide ion to form a precipitate of silver cyanide and a complex ion, $Ag(CN)_2^-$. Formulate the resulting overall equilibrium.

Solution:

Process	*Constant*
$Ag^+ + CN^- \rightleftarrows AgCN$ (solid)	$1/K_{sp}$
$Ag(CN)_2^- \rightleftarrows Ag^+ + 2CN^-$	K_i
$Ag(CN)_2^- \rightleftarrows CN^- + AgCN$ (solid)	K_i/K_{sp}

hence

$$K = \frac{[CN^-]}{[Ag(CN)_2^-]} = \frac{K_i}{K_{sp}}$$

K_s-K_{sp}. See K_a-K_s.

K_{sp}-K_{sp}. If two slightly soluble salts containing a common ion such as silver chloride and silver bromide are in equilibrium with a solution, the following simultaneous equilibria occur.

Process	*Constant*
(solid) $AgCl \rightleftarrows Ag^+ + Cl^-$	K_{sp_1}
$Ag^+ + Br^- \rightleftarrows AgBr$ (solid)	$1/K_{sp_2}$
(solid) $AgCl + Br^- \rightleftarrows Cl^- + AgBr$ (solid)	K_{sp_1}/K_{sp_2}

hence,

$$K = \frac{[Cl^-]}{[Br^-]} = \frac{K_{sp_1}}{K_{sp_2}}$$

Problem 11. If 0.1 *M* silver chloride is shaken with one liter of 0.1 *M* sodium bromide, what percentage of the sodium bromide will be converted to silver bromide?

Solution:

$$\frac{[Cl^-]}{[Br^-]} = \frac{1.6 \times 10^{-10}}{7.7 \times 10^{-13}} = \frac{208}{1}$$

hence 208/209 or 99.5 per cent of sodium bromide is converted.

OXIDATION-REDUCTION

Equilibrium constants for oxidation-reduction reactions are usually not made available in the tables of qualitative analysis texts but may be calculated from the single electrode potentials which are given, since, at 25°, the logarithm of K_r is equal to the difference between the two electrode potentials involved divided by $0.06/n$, where n is the number of electrons involved in the reaction. An important example of simultaneous equilibria involving oxidation-reduction is given by the oxidation of copper sulfide by nitric acid.

Process	*Constant*
(solid) $3CuS \rightleftarrows 3Cu^{++} + 3S^{--}$	K_{sp}^3
$3S^{--} + 8H_3O^+ + 2NO_3^- \rightleftarrows 12H_2O + 2NO(gas) + 3S(solid)$	K_r
(solid) $3CuS + 8H_3O^+ + 2NO_3^- \rightleftarrows 3Cu^{++} + 12H_2O + 2NO(gas) + 3S(solid)$	$K_{sp}^3 K_r$

hence,

$$K = \frac{[Cu^{++}]^3}{[H_3O^+]^8\,[NO_3^-]^2} = K_{sp}^3\,K_r$$

MORE THAN TWO EQUILIBRIA

The algebra of simultaneous equilibria which has been exemplified above may be extended to systems having more than two simultaneous equilibria. This is demonstrated by the following problems.

Problem 12. The solubility of hydrogen sulfide at 25° is approximately 0.1 *M*. Calculate the "solubility product" of hydrogen sulfide.

Solution:

Process	*Constant*
(gas) $H_2S \rightleftarrows H_2S$ (dissolved)	K_s
$H_2S + H_2O \rightleftarrows H_3O^+ + HS^-$	K_{a_1}
$HS^- + H_2O \rightleftarrows H_3O^+ + S^{--}$	K_{a_2}
(gas) $H_2S + 2H_2O \rightleftarrows 2H_3O^+ + S^{--}$	$K_s K_{a_1} K_{a_2}$

hence,

$$K = [H_3O^+]^2[S^{--}] = K_s K_{a_1} K_{a_2} = 0.1 \times 10^{-7} \times 10^{-15} = 10^{-21}$$

Problem 13. At what pH will zinc sulfide dissolve to form a 0.1 *M* Zn^{++} solution?

Solution:

Process	*Constant*
(solid) $ZnS \rightleftarrows Zn^{++} + S^{--}$	K_{sp}
$S^{--} + H_3O^+ \rightleftarrows H_2O + HS^-$	$1/K_{a_2}$
$HS^- + H_3O^+ \rightleftarrows H_2O + H_2S$	$1/K_{a_1}$
(dissolved) $H_2S \rightleftarrows H_2S$ (gas)	$1/K_s$
(solid) $ZnS + 2H_3O^+ \rightleftarrows Zn^{++} + 2H_2O + H_2S$ (gas)	$K_{sp}/K_{a_2}K_{a_1}K_s$

$$K = \frac{[Zn^{++}]}{[H_3O^+]^2} = \frac{K_{sp}}{K_{a2}K_{a_1}K_s}$$

$$[H_3O^+] = \sqrt{\frac{[Zn^{++}]K_{a_2}K_{a_1}K_s}{K_{sp}}} = \sqrt{\frac{0.1 \times 10^{-15} \times 10^{-7} \times 0.1}{1.2 \times 10^{-23}}} = 0.8\ M$$

pH = 1.1

Problem 14. Zinc hydroxide is equilibrated with 0.1 *M* sodium hydroxide. Calculate the concentration of zincate ion.

Solution:

Process	*Constant*
(solid) $Zn(OH)_2 \rightleftarrows Zn(OH)_2$ (dissolved)	K_s
$Zn(OH)_2 + 2H_2O \rightleftarrows 2H_3O^+ + ZnO_2^{--}$	$K_{a_1}K_{a_2}$
(solid) $Zn(OH)_2 + 2H_2O \rightleftarrows 2H_3O^+ + ZnO_2^{--}$	$K_s K_{a_1} K_{a_2}$

$$K = [H_3O^+]^2[ZnO_2^{--}] = K_s K_{a_1} K_{a_2} = 10^{-29}$$

$$[ZnO_2^{--}] = \frac{10^{-29}}{[H_3O^+]^2} = \frac{10^{-29}}{(10^{-13})^2} = 10^{-3}\ M$$

SUMMER PROGRAM IN THERMODYNAMICS

A SPECIAL summer program in the thermodynamics of equilibrium and non-equilibrium states will be given during the 1953 Summer Session from June 29 to July 10 at the Massachusetts Institute of Technology, according to Professor Ernest H. Huntress, Director of the M.I.T. Summer Session.

The program will mark the two hundredth anniversary of the birth in 1753 of Benjamin Thompson in Woburn, Massachusetts. Thompson, who as Count Rumford performed experiments on the generation of heat in the boring of cannon, was instrumental in discrediting the caloric theory and in stimulating the then-new theory of thermodynamics.

The summer program at M.I.T. will be designed to provide a sound exposition of classical thermodynamics and to explore the newer extensions of the subject into the kinetics of processes in which non-equilibrium states play an essential part.

Lectures on classical thermodynamics, kinetic theory and statistical mechanics, Gibbs' theory of equilibrium, and generalized thermodynamics and the kinetics of irreversible processes will be supplemented by a number of lectures and discussion sessions on special topics.

Four members of the M.I.T. faculty will give the principal lectures in the program: Joseph H. Keenan, Professor of Mechanical Engineering; Melvin A. Herlin, Assistant Professor of Physics; James A. Beattie, Professor of Physical Chemistry; and Laszlo Tisza, Associate Professor of Physics. Professor Lars Onsager, Professor of Theoretical Chemistry at Yale University, will be one of a number of special lecturers who will participate in the summer program.

Further information on this summer program and application forms may be obtained from the Director of the Summer Session, Room 3-107, Massachusetts Institute of Technology, Cambridge 39, Massachusetts.

THE ORGANIZATION OF SUBJECT MATTER IN ELEMENTARY ORGANIC CHEMISTRY

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ELEMENTARY organic chemistry, as taught in some colleges, involves a separate treatment of aliphatic and aromatic compounds. In our institution, the two types of materials are treated simultaneously according to a so-called "combined method." Teachers with some years of experience are familiar with the writings of such men as Cheronis (*1*) and Desha (*2*), each of whom has written a textbook using the "combined method" approach. It is doubtful that new instructors of organic chemistry consider the relative merits of the two methods when they first start teaching. It is more likely that they begin to teach according to the method to which they were exposed as students. This paper is written partly for the benefit of these young instructors and partly because the author feels the subject is of sufficient importance to warrant re-examination at reasonable intervals.

In 1929, Gilman, Heckert, and McCracken (*3*) wrote "The differences between aromatic and corresponding aliphatic compounds are largely of degree and not of kind. There are probably no rigidly exclusive aromatic characteristics." Some years earlier Henrich (*4*) wrote along similar lines: "The observed difference in the rate with which additions take place in the case of benzene and its derivatives as compared with unsaturated aliphatic compounds indicates a difference merely in degree. That it is not a difference in kind is supported by the fact that in certain types of aliphatic compounds the ethylenic linkage is just as unreactive as in benzene and that every variation between this and very reactive ethylene derivatives has been observed."

On the other hand some authors have made a considerable effort to develop the concept of "aromaticity." A good example of such writings may be found in the text by Fuson (*5*). Here the discussion is built largely around the observation that "The structural characteristic that all aromatic rings have in common is the so-called sextet." The subject matter is developed very convincingly and the concept of "aromaticity" seems to be a useful one. However, at the start of his chapter the author makes a statement which may well cause one to be very doubtful of the value of emphasizing "aromatic character" to the students of the elementary course. Particularly is this so when such emphasis leads to the arbitrary separation of the course into aliphatic and aromatic divisions. The statement is as follows: "The line of demarcation between aromatic and aliphatic compounds is so ill-defined that no simple definition of aromatic character or aromaticity has been agreed upon. Perhaps the outstanding characteristic of aromatic compounds is the peculiarly diminished reactivity of the double bonds. Although benzene is definitely unsaturated, its unsaturation is of a lower order than that of open-chain olefinic compounds." It is of more than passing interest to note that in the elementary text by Fuson and Snyder (*6*), aliphatic and aromatic compounds are treated simultaneously.

The passing of the years, with the accumulation of ever-increasing amounts of new data, serves only to confirm the views expressed in references (*3*) and (*4*) and, in part, in reference (*5*). Yet, at the present time, even though there may be a great deal of agreement with these views, nevertheless we find the majority of college professors reluctant about combining aliphatic and aromatic compounds in teaching the first course in organic chemistry. The "orthodox" method of presentation, involving the separation of aliphatic and aromatic compounds, has certain good features but these advantages are more than offset by the disadvantages inherent in the system. When taught this way, all too often the student fails to see the subject broadly; he has the course neatly tied in packages, two of which are labeled "aliphatic" and "aromatic." To demonstrate this point ask a student who has recently completed the course to write an account of the effect of halogens on hydrocarbons. If he is a product of the "orthodox" course he is apt to show first the halogenation of alkanes, then alkenes and alkynes. He will describe next, and as something apart, the behavior in benzene. He treats this hydrocarbon as something different but seldom notes the difference is only one of degree.

It is assumed, in the "orthodox" course, that by the end of the year the student will have, automatically or otherwise, integrated the subject matter of the course and will have recognized that the separation into aliphatic and aromatic divisions was a pedagogical device employed to "simplify" the presentation. Unhappily, this integration does not appear to occur in many cases and the student persists in thinking in terms of aliphatic and aromatic chemistry rather than in the more general terms of organic chemistry.

From a logical viewpoint it is hard to argue against the simultaneous treatment of aliphatic and aromatic compounds. Not only does the "combined" approach treat organic chemistry as it is known to be, but in so doing it gives the student a broader picture of the subject. Furthermore, one of the very best ways to stim-

ulate interest in the work is to give him an early introduction to the fascinating chemistry of benzene. Then too, by avoiding duplication of effort, not possible in the "orthodox" presentation, extra time is gained for the inclusion of new material. This point is of no small significance in view of the rapidly increasing volume of material in the field of organic chemistry; an extra two weeks is a welcome addition to the course.

Perhaps the most undesirable feature about the "combined" method of teaching is the lack of texts written in this way. At the present time the instructor is limited to, not necessarily the "best" book, but the most suitable one using his style of presentation. Since the trend toward the combined method seems established, though moving slowly, time or an acceleration in the rate of adopting the method should provide an increased number of authors interested in the style.

The remainder of this paper will be devoted to short discussions of a selected number of topics as they are presented by some professors using the "combined" approach. It is hoped the discussion will serve to remove or minimize some of the pedagogical difficulties attributed to the method.

The Structure of Benzene. During the study of alkanes and their derivatives the student is introduced to the concept of resonance by using the nitroalkanes as examples. This is a relatively simple way to approach a troublesome subject. Reaction mechanisms, involving electronic interpretations, are used as the student progresses from the alkanes to the alkenes and alkynes and then dialkenes. After a description of 1,3-butadiene the discussion soon leads naturally into an account of resonance in the benzene ring. By this time the student is reasonably conversant with the concept and benzene provides an especially interesting example of it. If electronic interpretations have been emphasized to this point then the student is quite able, also, to grasp the ideas on orientation in the ring. He has, in the meantime, acquired a sufficient knowledge of organic reactions to appreciate the other ideas usually presented in support of the structure of benzene.

Halogenation. Benzene resembles both alkanes and alkenes in that it reacts by both substitution and addition. It is of interest to note that certain alkenes also undergo both types of reaction, particularly in the case of chlorinations. A common example of the latter type is the high temperature chlorination of propene to allyl chloride; another example is the low temperature chlorination of isobutylene to methallyl chloride. It is not a difficult step for the student to go from a consideration of the chlorination of alkanes and alkenes to a consideration of the chlorination of benzene. The study of the chemistry of benzyl chloride fits in well after a discussion of allyl chloride. Although experimental conditions and reaction mechanisms often may differ considerably, these differences tend to make the presentation more interesting and not necessarily more difficult.

Nitration and Sulfonation. After having observed the behavior of alkanes and unsaturated compounds with nitric acid and with sulfuric acid, a comparison may be made with the behavior of benzene toward these reagents. The direct nitration of benzene may be compared with the direct nitration of tertiary alkanes or with isobutylene. The high temperature nitration of propene also may be considered at this point. In a similar fashion the preparation and properties of benzene sulfonic acid and the sulfonic acid formed from isobutane may be compared. The sulfates formed with alkenes serve to emphasize the greater reactivity of most alkenes over benzene in addition-type reactions.

Friedel and Crafts Reaction. This reaction is thought of, usually, as an "aromatic" reaction; however, it is also a reaction involving alkyl halides and a discussion of it falls naturally into an early spot in the course if the chemistry of benzene has been considered.

Cannizzaro Reaction. This reaction, often illustrated with benzaldehyde has, of course, its counterpart in formaldehyde and certain substituted aliphatic aldehydes.

Ozonization. Alkenes and benzene are very similar in their behavior toward ozone. If the reaction has been illustrated with alkenes it has considerable significance to the student if he views it again very shortly in connection with the work on the structure of benzene.

Wurtz Reaction. This reaction has its aromatic counterpart in the Wurtz-Fittig reaction and the two, logically, are considered together.

Among miscellaneous items adding early interest to the "combined" course may be mentioned the following:

(1) The conversion of heptane to toluene.

(2) The hydrogenation of benzene to cyclohexane or the dehydrogenation of the latter compound to benzene.

(3) The formation of benzene from acetylene.

How is the "combined" approach applied to the study of various hydrocarbon derivatives? In the case of alcohols and phenols, for example, it seems better to study these two materials together under the heading of hydroxy compounds than to separate them into alcohols one semester and alcohols and phenols the second semester. Students are strongly attracted by the similarities and differences in properties among the various alcohols and phenols. The fact that the behavior of the phenolic hydroxy group is strongly influenced by the nature of the phenyl group adds much interest to the discussion and again does not increase its difficulty. This is particularly true if, previously, emphasis has been placed on the effect of substituents of various groups, both aliphatic and aromatic. Note that the greater emphasis is placed on the behavior of the functional group and the lesser emphasis on the nature of the rest of the molecule, without discounting the importance of the latter.

An examination of compounds with other functional

groups leads to similar conclusions. Why not consider the behavior of chlorobenzene shortly after a consideration of vinyl chloride; aniline with hexylamine; benzoic acid with acetic acid, etc?

The writer has been teaching the "combined" course for a number of years and has consulted with other professors who have been teaching it for longer periods of time. As a result of this experience he is of the opinion that the simultaneous treatment of aliphatic and aromatic compounds in the elementary course is a highly satisfactory way to teach the course and that there are few if any, pedagogical difficulties involved in the process which are not easily overcome. When compared with the "orthodox" method, this style of presentation is not only a more logical way to handle the subject but is probably a more interesting way from the viewpoint of both the student and the professor.

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UNIVERSITY INSTRUCTION IN CHEMICAL LITERATURE

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The need for training chemistry students in making use of the literature of their field was pointed out as early as 1916 by Bacon, *et al.*, in a report to the Committee of 100 on Scientific Research.[1] A need thus existed then, and this need is certainly more urgent today, when there is a vastly more voluminous literature, resulting in added complexity of subject matter.

There are several ways of introducing students to the use of the literature. Budington[2] described several methods used in teaching engineering students, with whom the problem is essentially the same as with chemistry students, or, for that matter, with students of any science depending largely upon the serials literature.

If we eliminate freshman library orientation, which, although useful, comes at too early a stage of a student's training, we have the following possibilities:

(1) Giving occasional talks on library and literature technique to classes.

(2) Setting aside one or more periods in a special course and devoting it to instruction in the literature.

(3) Devoting an entire course to chemical bibliography.

We have tried the first two methods, and have come to the conclusion that only the last-mentioned one gives the student sufficient training. Guest lecturing in one course is not successful, since most of the instructors have more material than time to cover it, and are reluctant to give up any of their periods. Making library instruction a part of a special course, such as a senior seminar, is not the solution, since in the two to four lectures devoted to the literature, only a small part of the necessary knowledge can be covered. It takes at least one hour to explain the classification system, card catalogue, and correct bibliographic citation, and if one hour each is spent on *Chemical Abstracts*, *Beilstein*, and *Gmelin*, there is no time left to cover any tools in analytical chemistry, physical chemistry, or biochemistry, not to mention applied chemistry or other tools in organic and inorganic chemistry. Lectures of this type tend to be cramped, and, hence, students get very little from them.

To obtain some idea of how other major universities solve this problem, we sent questionnaires to 75 schools listed as granting the Ph.D. in chemistry in 1948–49,

[1] Bacon, R. F., C. E. K. Mees, M. C. Whitaker, W. R. Whitney, and W. H. Walker, *Science*, **45**, 37 (1917).

[2] Budington, W. S., *Coll. Res. Lib.*, **12**, 268–72 (1951).

in a report published in *Chemical and Engineering News*.[3] There are 76 schools listed, the University of Wisconsin being the 76th school of the present study.

The following questions were asked:

1. Does your school give a course in chemical literature?
2. Is it a required course?
3. Is it given for credit?
4. Is it taught by the librarian?
5. Does the librarian give occasional lectures on the use of the library or literature?

All questionnaires were addressed to the librarians of the chemistry departments. Sixty questionnaires, or 80 per cent of the total, were returned, some of them with comments which will be discussed below. The statistics have been compiled on the basis of these 60 schools.

Courses. Thirty-two schools offer formal courses in chemical literature. Of these, 20 are required courses. The requirement is broken down by some schools: two required such a course for undergraduates, five did so for graduates, and two did so for both undergraduates and graduates. Eleven schools did not break down the requirement with regard to graduates and undergraduates.

Credits. Fifteen courses carry one credit; ten courses carry two credits; one course carries three credits, and six schools did not give the number of credits. When there was a choice of credits, the lower number was calculated, with the exception of one zero- or one-credit course, where one credit was used.

Instructors. Twenty-four courses were taught by members of the department other than librarians. Eight courses were taught by librarians. If this question was not answered completely we assumed that the librarian did not teach the course. In five schools offering no formal courses, the librarian gave occasional talks on the literature. In six schools offering courses not taught by the librarian, librarians gave lectures on the literature or library. In five schools, librarians taught formal courses and gave lectures in addition to these courses.

Comments on the last two questions seem to be in order. We are of the opinion that the librarian, providing he has the proper chemistry background, is best qualified to teach a course in chemical literature. The librarian is in constant contact with the literature and has had formal instruction as well as everyday informal instruction through answering reference questions, searching the literature, and continuous use of the literature, in short, the background in the very field in which the student will have to be instructed. Instructors know more about their special fields, but librarians, being in continuous contact with all the literature of chemistry, and, probably, with the literature in closely related fields, have a better general overall view of the field.

Unfortunately, the requirements were not broken down sufficiently, but to judge from Budington's study and from the situation at the University of Wisconsin, it would appear that the junior year would be the most appropriate time to give this course. Students are then well along in their major, having had a year of organic chemistry and probably being in process of taking physical chemistry. For graduate students without previous training in the literature and in need of such training, the first semester is best. In neither case should this course be given merely for the sake of having extra credit, which no school seems to need. To avoid requiring students to take the course who have taught themselves the use of the literature, a test on their knowledge could be given previous to registration in such a course.

Two types of comments were enlightening. The majority of schools elaborating on their course explained that it consisted of lectures followed up by written assignments, application of course material in the library. Students in one school complained that they were overworked for a two-credit course, and three schools had once had courses or series of lectures, but these were dropped because of lack of interest. It is not surprising, in a crowded curriculum, that very few students will volunteer for extra work even though it might benefit them. The chances are that students who volunteer need the instruction least. These comments indicate that courses in the literature, in order to accomplish their purpose, have to be required or "strongly recommended."

SUMMARY

The need for formal training in the literature of chemistry is becoming more important with the increasing complexity of, and the student dependence upon, the literature. Of 60 schools offering the Ph.D. in chemistry, 32 give formal courses in chemical literature, 20 of which are required for chemistry majors. These courses run for one semester and are given for one to three credits. Due to the reluctance of students to assume extra work on a voluntary basis, it would seem that required courses in the junior year, or the first year of graduate school, are most effective in teaching students the use of the literature. Students who are sufficiently familiar with the literature should be exempted from the requirement. Single lectures and even two to four lectures in a special course such as the senior seminar proved to be unsuccessful at Wisconsin, since the student can only be given a small portion of what he should know about the literature, and that small portion necessarily out of context. The most efficient way of teaching the use of the literature in a formal course is to explain the function of each type of material and to give assignments of practical literature problems, which make use of the various tools taught.

Students take on the average of 40 credits of chemistry in their undergraduate work. Of these 40 credits, there should be room for one or two credits to teach them where to find the material they must use throughout their professional careers.

[3] Progress Report No. 21 of the Committee on Professional Training, *Chem. Eng. News*, **28**, 770 (1950).

AXEL FREDRIK CRONSTEDT[1]

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Two hundred years ago chemical knowledge consisted of a heterogeneous body of facts related to the very few elements then known and the somewhat larger number of compounds which were obtained in sufficiently pure form to be characterized. Much of this information was applied to practical applications in medicine and to the various industrial arts. Robert Boyle had stated his gas laws: nevertheless theories about matter, its ultimate form, combustion or attraction of one type of substance for another, were either nebulous or inaccurate. To create a system of chemistry with logical interrelationships, accurate qualitative and quantitative analysis, by which a considerable number of pure substances might be identified and incorporated into a classified whole, had to be developed.

Several factors stimulated the accomplishments of the eighteenth century Swedish analysts who laid the foundations for the influential period of their nineteenth century successors, when Berzelius became the arbiter of chemistry in western Europe, a post which ruled the chemical world. Universities had been founded at Uppsala in 1477 and at Lund in 1668, where the libraries had the right to receive everything published in the country. At Uppsala the renowned Linneaus lived in the botanical garden. An observatory had been erected in 1730. Any students at that university could have excellent training for that time. The Swedish Royal Academy of Science, which offered in its journal an outlet for exchange of ideas for those interested in scientific study, was established in 1710. Although the political atmosphere in Sweden was not stable, the country was distant from the warring armies of the European continent. The natural resources of Sweden were remarkable for timber and mineral deposits of great potential value. The iron ore alone represented some of the richest in the world. In the eighteenth century Sweden became the greatest exporter of iron products of all the iron producing countries. Although she lost this position when coal was substituted for charcoal as a reducing agent, the quality of steel products for cutting tools remains superb.

Axel Fredrik Cronstedt, destined to be one of the able analysts who enriched chemical knowledge, was born on December 23, 1722, in the province of Sodermanland. He was the child by a second marriage of a lieutenant-general, Gabriel Cronstedt, who was able to supervise his son's studies and to employ a tutor for him from 1726 to 1738. His mother was Maria Elizabeth Adlersberg, the daughter of an assessor. Cronstedt's instruction was continued at Uppsala where he listened to lectures as a nonregistered student. His most influential instructors were Andreas Celsius the astronomer and devisor of the thermometric scale, J. G. Wallerius, a chemist, and Daniel Tilas, a geologist. Although his father hoped that Cronstedt would follow in his own military footsteps, the young man had a decided talent for natural science and for drawing. In the mineralogical chemical laboratory of Wallerius, Cronstedt met Sven Rinman who encouraged him to study chemistry. Since Cronstedt did not have enough money to go to the University, he entered the School of Mines, where Tilas was his best counselor. By 1742 he was employed to do some practical teaching on a temporary basis, but his work and his education were interrupted several times and it was some years before he completed his studies.

MINING AND METALLURGY

In 1743 the peasants moved into the capital to obtain their rights, and the Russian Navy became a menace on the Baltic Sea. Since the political situation in Sweden was somewhat precarious, General Cronstedt, who was commandant of the fortifications at Stockholm and the neighboring archipelago, and who had no money from the government to hire a secretary when he made a tour of inspection of these fortifications, asked the school to release his son. Cronstedt left the school in August, 1743, and traveled as his father's secretary for three months on the east and west coasts of Sweden, where he saw his first mine in November, 1743. Many have thought it was the turning point of his life. But he was not dressed for a descent into the mine and thus saw little. It was in 1744 that he had a real opportunity to study metallurgical operations. That summer, with the recommendation of the School of Mines, Cronstedt traveled with G. G. Reuterholm to visit the mining country. The next year, Tilas advised him to go on another tour to study the production of silver and lead and the relationships of the miners with their management. This year, when Cronstedt took the place of Alexander Funk, a friend of Tilas, who was engaged in making typographical maps of the mining areas, Cronstedt made several personal contributions to the project. When Funk and Tilas quarreled over the map work, Funk resumed his position and Cronstedt went back to the School of Mines to take a course in assaying and chemistry under Georg Brandt, who was the discoverer of cobalt, the fifteenth element to be identified and the fifth element to be isolated in historic times. Brandt had traveled to Leyden to hear the lectures of

[1] Presented before the Division of History of Chemistry at the 121st Meeting of the American Chemical Society, Buffalo, March, 1952.

Hermann Boerhaave, the most eminent chemist of his day and had taken his medical degree at Reims. After a sojourn in the Hartz Mountains where he learned all he could about metallurgy, Brandt had returned to Stockholm and the chemical laboratory of the School of Mines.

In the summer of 1746, when Cronstedt surveyed copper mines, he became well versed in the extraction of that metal. Meanwhile, he began his extensive and valuable mineral collection. That same year he had an offer to go to Tunisia to build and operate a copper mine. Before he started, to his great disappointment word came that Tunis and Algeria were at war; so he was unable to go. In the last months of 1746 Cronstedt accompanied Tilas as amanuensis on further map-making trips. As a temporary measure, Cronstedt was put in charge of the library of the School of Mines and had the responsibility of preparing the institution for the royal inspection of King Adolph Fredrik. Further trips were made with Tilas to copper mines and to map the entire island of Singön. This map, which might have had historical interest, was lost in a fire when Tilas' house burned in 1751. Since Tilas was an excellent geologist with considerable ability to identify minerals, he taught his young protégé much. He urged him to "honestly observe and tell the truth as can be done in the mining science."

In 1748, when Cronstedt had finished his studies, a short-lived mining and geological company was formed with Tilas, Funk, and Rinman. The fruits of this organization for Cronstedt were the friendships formed and the correspondence between the members. As a result of his trips and investigations from 1748 to 1753, Cronstedt compiled an account of the "Mineral History of Westmanland and Dalecarlia." This volume, modeled after a book by Tilas on the "Mineral History of Finland," became a standard for its successors. In 1754, Cronstedt, Rinman, and von Swab were made members of the Swedish Academy.

From 1755, Cronstedt had a home in Skisshyttan, a somewhat isolated place where he supervised a silver mine. In the house he had a good assay room in which, between his trips, he studied chemistry and mineralogy. Reports from these studies were presented before the Swedish Academy. In the winter he often went to Stockholm, especially those seasons of 1751–52 and 1755–56 when he was a member of the Parliament. Because of his known skills, his zeal, and his enthusiasm for his work, he was made a member of committees which studied mining, merchandizing, and economics.

In 1752, with Anton von Swab, Cronstedt inspected the Swedish calamine mines to determine whether the native ore could be substituted for the imported ore to produce brass. Rinman continued these experiments after Cronstedt had done considerable work on the washing, oxidation, decomposition, and extracting of the calamine.

The next few years were varied. Cronstedt went to Norway for six months, where he increased his mineral collection. With von Swab, he studied the silver mines at Stora Kluken, the minerals in Jamtland, and various deposits of rock crystal and limestone. By January, 1757, he had returned to Skisshyttan for a much needed rest. Twice he was ill with the ague and once with the so-called Uppsala fever. Meanwhile, he was working on a manuscript of the greatest importance for which he is most famous. This was his mineral classification, an epoch-making revolution in mineralogy. The system was based upon chemical composition instead of the time-honored superficial outer characteristics of form, color, and hardness. This idea, which was expressed in a letter to Tilas in 1753, Cronstedt himself said was encouraged by von Swab. Earlier, a German chemist, Pott, considered a similar scheme but did not develop it. The first draft of Cronstedt's mineralogy was sent to Rinman in 1756, and by 1757 he had completed his manuscript. On May 12 of that year Cronstedt wrote to Tilas that he was not planning to publish his ideas. However, when Cronstedt's physician saw the written document he persuaded him to have it printed anonymously. Two brothers, Peter Jonas Bergius and Bengt Bergius, supplied the needed money.

In 1756, when Cronstedt had hoped to receive an appointment at the School of Mines, he was passed over in favor of a younger man. Cronstedt complained bitterly that he should have had this secretaryship since he had "spent the time, money, and health to no avail . . . were they not qualifications for being put into the office?" In 1758 he obtained a position in Norway to work in a silver mine near Jarlsberg, but von Swab and Tilas persuaded him to postpone his departure for three months. During this time Cronstedt was appointed superintendent of mining operations in the "Oster and Västerbergslagen," a post which he accepted. Until his marriage, which took place in Ängelsberg, Västervåla, in 1760, he lived in the superintendent's house. His wife, the daughter of an army colonel, Lorens Nicklas Söderhielm, was Gertrud Charlotta Söderhielm, who was born in 1728 and died in 1769.

LATER LIFE

After 1757, and more markedly after his marriage, Cronstedt turned to the study of botany, in which he became interested from the practical point of view. He wrote to Tilas, in connection with his manuscript on mineralogy, that his recreation in the country was now botany, since the mineral kingdom had proved to be a disappointment because he no longer had the money or the courage to continue its studies. Mineralogy seemed to him inflexible and without thanks. When he had completed his distinguished classification his inclination to study plants began to take precedence over all else in his spare hours.

Before his death Cronstedt had acquired considerable property, some of which came from his wife. He had a

very well-equipped laboratory and a large library in his own home at Nysshytte. The constant duties of his last years allowed him little time for scientific pursuits, as he had become an advisor for many who had mining interests. The lack of freedom was so wearing that he complained that it was "impossible to do chemical work." He seldom went to Stockholm except when he was returned to Parliament in 1765. Again he was made a member of those committees on which he could best serve. In the summer of 1765, only five years after his marriage, he returned home from Stockholm exhausted and ill, to die in a few days on the 19th of August. Not yet 43, Cronstedt had become an eminent mining expert who might have made many further contributions if he had lived.

The library of the Swedish Royal Academy owns some 50 volumes of manuscripts which contain Cronstedt's writings and his correspondence. More of his letters are located in the Royal Archives, the School of Mines, and the Royal Library. The mineral collection of approximately 3000 items, which was left to his son, was taken by his widow to her new home after her remarriage to Captain G. G. Reuterholm where it remained until 1803. In 1753, Cronstedt had constructed special equipment for the cutting and polishing of the minerals so that he obtained a better conception of the crystal structure. He was anxious to show that his native country had as beautiful types as any other land. When the Queen, Luisa Ulrika, expressed an interest in his specimens, he also did some collecting for her. When the School of Mines heard that the collection was purchased by a wealthy Danish landowner and collector who lived in Paris, it tried, without success, to persuade the king to prevent its removal from Sweden. A very prominent Swedish chemist, Peter Jacob Hjelm, the discoverer of molybdenum, was asked for his opinion. Lacking appreciation of Cronstedt and not recognizing the value of the collections, Hjelm's advice was to let it go. His excuse was the lack of good labels on the specimens. Nevertheless, Cronstedt's personal catalogue of the collection was known to be in the hands of the Dane. Hjelm must have been careless in his judgment. What happened to the collection after it reached Paris is not known. Perhaps it was sold again. The troubled state of France in the Napoleonic era made it difficult to trace its fate.

MINERAL CLASSIFICATION

Cronstedt's scientific contributions were made in a relatively short period. The great geologist, Abraham Gottlob Werner, of Freiberg, wrote that the "Essay on the New Mineralogy," as his classification was called, should give to Cronstedt the honor of being called the father of that science. In the preface of this book printed in 1758, which was the turning point in the history of mineralogy, we find Cronstedt's views. He believed that while men in the preceding century had shown great appreciation for the study of philosophy and antiquity, the time was now ripe for a better understanding of science. An adequate classification of minerals was needed because previous attempts to classify them had been based only on physical properties and not chemical composition. Since Cronstedt felt that previous scholars had assumed unnatural relationships, he determined to organize his ideas into an improved system which would retain the good ideas and reject the bad ones in the older systems.

The new system comprised four classes, earths, bitumens, salts, and metals. The definitions of the classes showed the influence of comtemporary belief. The *earths* could not be flattened out, did not loose their substance under a hammer, were for the most part insoluble in water and oil and retained their shape when ignited. The *bitumens* burned in fire, and dissolved in oil but not water. The *salts* usually dissolved in water, to which they gave a flavor and from which they crystallized when the water was evaporated. The *metals* were the heaviest known bodies, which were in part malleable and in part reducible to their principles according to the language of phlogiston. By restoring the inflammable principle which they lost by ignition, they were returned to their original form. The *earths* had nine divisions, calcareous, siliceous, granites, clays, mica, fluorspar, asbestos, zeolite, and magma. The *bitumens* were amber, petroleum, asphalt, and the phlogistic material, sulfur. Salts of vitriolic acid, marine acid, the alkali minerals, and borax were grouped together. The half-metals which could not be found free in nature were classed with the metals. The last lines in the book were: "I await with pleasure the judgment of those who know this type of work and have the same sentiments. I shall be ready to receive the judgment that readers form for this essay." Cronstedt's astute opinions about many minerals are too numerous to mention. Although there were errors according to present views, the value of his chemical classification is undeniable.

Like all innovations, the new classification was not acceptable to the conservative; von Swab of course approved. Tilas, since he had helped another of his pupils, Wallerius, to organize a mineral system, was not quite satisfied. After Cronstedt's death, Linneaus, in the 12th edition of his "Systema Naturae," published in 1768, finally expressed approbation, although he was inclined to other directions.

Linneas thought that the classification was the voice of von Swab but the hand of Cronstedt. Another mineralogist, von Engestrom, who was a pupil of both von Swab and Cronstedt asserted that Cronstedt was the real author of the new mineral system and had an original manuscript available at that time to prove it. An incomplete German edition was soon used as a text at the Freiberg Mining academy, which was the best in Europe. With its success assured, 16 years after Cronstedt's death, a second edition was printed with considerable profit.

DISCOVERY OF NICKEL

As the discoverer of a new half-metal, which he named nickel, Cronstedt is best known to chemists. Nickel had been used in alloys by the Chinese without identification. In 1694, Hairne had found an odd mineral which resembled copper ores in color. Vain efforts to produce copper from this substance lead to its being given the insulting name of kupfernickel. In 1726 a paper in the *Philosophical Transactions* reported that this material contained cobalt and copper. Three years later, John Andreas Cramer, a German miner, suggested cobalt alone. Linneaus specified copper and arsenic and Wallerius cobalt, iron, and arsenic. Cronstedt, trained in Brandt's laboratory, did not follow the prevalent custom of obtaining percentages of constituents and then making them up to 100 per cent; thus hiding errors which might otherwise reveal new substances. His more careful study of a similar-appearing earth found in a cobalt mine in Helsingland was reported to the Swedish Academy in 1751. The following facts were presented as conclusive evidence of the presence of a new elementary substance. Action of air on the ore produced a green earth. This when treated with vitriolic acid gave a green vitriol, which changed to a gray *colcothar*, or oxide in modern terms, when strongly heated. A hard, white metal was obtained by reduction which gave no test for copper with borax, nitric acid, or alkali. Further analysis indicated the presence of a new element and not merely some cobalt "which had lost its soul." In addition, Cronstedt prepared the calx, the sulfide, and a copper alloy of the element, which he named "nickel." Every known means was used to verify his conclusions.

Although this report of Cronstedt's was accepted by many chemists, others refused to agree. Two Frenchmen—Sage in his "Elements of Local Mineralogy," printed in 1772, and Monnet in his "Treatise on the Solution of Metals," published in 1775—assured their readers that cobalt and nickel were actually the same. In the latter year the problem was examined in a masterful manner by Torbern Bergman who proved that Cronstedt's nickel had been somewhat contaminated. With great care, Bergman himself prepared a pure form that was attracted by a magnet and showed in addition all the characteristics of a new element similar to iron. Doubts were not completely settled until 1811, after the investigations of Proust (1803 and 1806), Richter (1804), and Tupputi (1811), who between them isolated and distinguished the nickel salts as new entities. Sixty years is a long time to wait for vindication but chemistry moved slowly and conservatively in those days. For many years the European chemists knew no use for nickel. Cronstedt, as early an 1754, had sought to use the nickel calx as a paint pigment to no advantage. Thenard, as late as 1824, wrote in his "Treatise of Chemistry" that nickel had no practical importance. Scientific advancement has changed that view.

USE OF BLOWPIPE

The painstaking analytical work accomplished by Cronstedt was carried out before the present-day systematic procedures and modern inventions. The reagents which he had at hand were the few inorganic acids, bases, and salts which were known in the Middle Ages. The best instrument was the balance. Various vessels for studying such operations as solution, evaporation, distillation, sublimation, fusion, and ignition had been devised. The records of Johann Heinrich Pott about 1745 tell effectively the type of examinations which were made. Pott described the behavior of earths and stones which he recognized by the appearance produced either alone or in combination with other bodies or with different salts or glasses in different proportions and at different temperatures. Melting points could be ascertained and found to be reproducible for pure substances. Pott learned that clay did not melt at the highest temperature which he could reach. Its fusibility with the addition of chalk, borax, fluorspar, lead calx, or gypsum gave a variety of appearances to the molten masses, dependent upon the quantity of additional material used. The idea of using a blowpipe to detect very small amounts of materials was the next logical step.

It is doubtful to whom the honor belongs of introducing the blowpipe as an implement for chemical analysis. In his volume on "The Use of the Blowpipe in Chemistry and Mineralogy," Berzelius recorded the history of the device. Previously, the blowpipe had been used in the arts before it was adopted by the chemists. Such a tool had been utilized in the process of soldering by goldsmiths, who by its use could force the flame of a candle or a lamp against any special point they wished to heat. It enabled them to solder almost any type of jewelry without heating more than a small part of the object. The Venetian glassmakers had learned to blow molten glass into small molds. Libavius described such glass products in his chemical text in 1613. In 1660 the records of the Academia del Cimento mentioned the use of glass tubes as bellows to construct fine glass objects. In 1669 Kunckel wrote that a double bellows for a blast lamp would be advantageous for the chemist who wished to reduce a metal calx. By means of the blowpipe the flame could be directed upon the calx, which was placed in the hollow of a charcoal block. The first man to carry out such operations extensively was Cramer, who was able to melt small bits of metallic material in this manner. By observation of this process, he obtained a quick proof of the composition. Cramer only considered the general appearance, the melt, and the result when a little borax was added. Because blowing with the mouth was so difficult and also thought hazardous to health, bellows were designed and described to produce the air blast.

The blowpipe was introduced into Sweden for special chemical analysis by one of three men. In 1746 Sven Rinman used the instrument to investigate an iron-tin

earth which he subjected to liquation. Bergman stated that von Swab performed blowpipe tests on minerals in 1738. Since von Swab published nothing on the subject it cannot be ascertained just how extensive his research might have been. The first written record describing work with an antimony regulus appeared in 1748. Von Engestrom insisted that Cronstedt was the first man to think of the use of the blowpipe to investigate minerals and emphatically stated that von Swab acknowledged that fact. While it is difficult to prove the truth, it is certain that Cronstedt was, if not the first, one of the first to perfect the blowpipe technique, which he clearly described in his search for nickel. By employing fusible reagents and by noting the changes produced by these fluxes Cronstedt judged with certainty the composition of minerals. When established, this method enabled the investigators to detect very small differences in composition in a simple and accurate manner. Upon this improved analysis Cronstedt based his system of mineralogy.

During these years when means of communication were less reliable and less rapid, Cronstedt worked almost alone. He attained a high degree of ability in the use of the blowpipe before anyone outside his small sphere knew what he had accomplished. When Von Engestrom published the first English edition of the mineralogy, he added a chapter on the blowpipe which described Cronstedt's methods for manipulating this instrument. When the English work was brought back into Sweden it aroused such general interest that it was translated into German, Italian, French, and Russian. At first men were unwilling to learn this difficult art until they were convinced that it was useful. For this reason the use of the blowpipe was confined to Sweden, where his contemporaries had seen Cronstedt work and could imitate him. Improvements by Bergman made the blowpipe indispensable. Gahn, Bergman's successor, became still more proficient. Berzelius, whom Gahn taught, said, "I saw him (Gahn) obtain with the blowpipe distinct particles of metallic copper from different sorts of paper after burning a quarter of a sheet to ashes." Gahn never published an account of his refinements. Berzelius watched Gahn and recorded in his "Elements of Chemistry" what he observed.

The simple blowpipe of Cronstedt, whose preeminence in the art of its use was acknowledged by all, was modified by many men, among whom were Gahn, Voigt, Tennant, and Wollaston. Later instruments were made of silver or tinned iron with a brass point. The flame was produced by a lamp. For the portable equipment designed by Cronstedt, a candle was substituted for the lamp. The charcoal support prepared from pine or other soft wood was modified by the use of platine or mica. Cronstedt used three general reagents, basic carbonate, sodium borate, and sodium ammonium phosphate. It was fortunate that at the outset of blowpipe analysis the three best reagents had been found. In the fourth edition of Berzelius' work on the blowpipe the number of reagents had been increased to 19. However, five of them were used to identify elements unknown to Cronstedt. The basis which he built had a solid structure, which was developed very slowly for the next 75 years.

OTHER STUDIES

In addition to the two papers on nickel which were published in the Swedish Academy *Transactions*, Cronstedt wrote six others. Many indicated his interest in resources which would be of economic use to his country if they were available. In 1753 he was the author of a paper on gypsum. He wrote that "although Sweden is blessed by God in having an abundance of minerals, nevertheless there is a dearth of gypsum, or perhaps it has not been discovered in sufficient quantity. Its value is dependent upon the unique property that makes it harden in water after ignition."

Cronstedt made a brief report on a mineral water from which silver could be precipitated. He was unwilling to state just how this was possible, but such waters should indicate the composition of the rocky material from whence they came. He investigated the blue color which he could obtain from a variety of grass known commonly as cow wheat or *melampyrum* by the botanists. Although he hoped that he might find a substitute for indigo, the blue color was too transient. Asked by the Royal Academy to study the usefulness of an iron hearth in a bake oven, he found that it saved two thirds the amount of wood required for one made of brick. He noted the expansion of the iron when heated, which might crack the surrounding walls; so care must be exercised in its construction. Cronstedt deplored the destruction of the Swedish forests by fire, which reduced the quantity and quality of building materials for homes and ships and also the charcoal for production of iron. He studied the use of tile, brick, and slate as substitutes for housing construction to relieve the shortage. This led to an economic survey of slate deposits, clays for ceramic wares, and the highways which were required for transportation of the finished products. He was aware of the loss of heat in the operation of the iron furnaces and discussed the problems of finding ways to utilize it. In a long article he published a mineral history of Jamtland, which was a costly but important inspection to discover the available natural resources in that district. In addition, Cronstedt pointed out that there should be amicable relationships between the agricultural and mining interests in the country. Many of the opinions expressed in these papers seem almost contemporary in spirit.

The links in the chain of investigators in which he is a part are Boerhaave, Brandt, Cronstedt, Bergman, Gahn and Berzelius. For 150 years the eminence of the Swedish School was maintained without interruption. In his relatively short scientific life, Cronstedt's work was so superior that it became the model for Bergman, who has been called the "father of analytical chemistry." Cronstedt himself might well be given that title. The discovery of a new element, the introduction of new and improved analytical techniques, classification of minerals, and practical applications of chemical information

obtained in the laboratory were Cronstedt's main contributions. His memory is perpetuated by a silicate named crondstedtite in his honor. What can be said of him can be said of some research workers today. Years ahead of his contemporaries, he was somewhat misunderstood in his lifetime, because other men did not grasp the import of his conclusions. Cronstedt represents the skilled research man of any age, intensely curious, who is devoted to finding the truth for the sake of science and his country.

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AN ELECTRONIC INTERVAL MARKER

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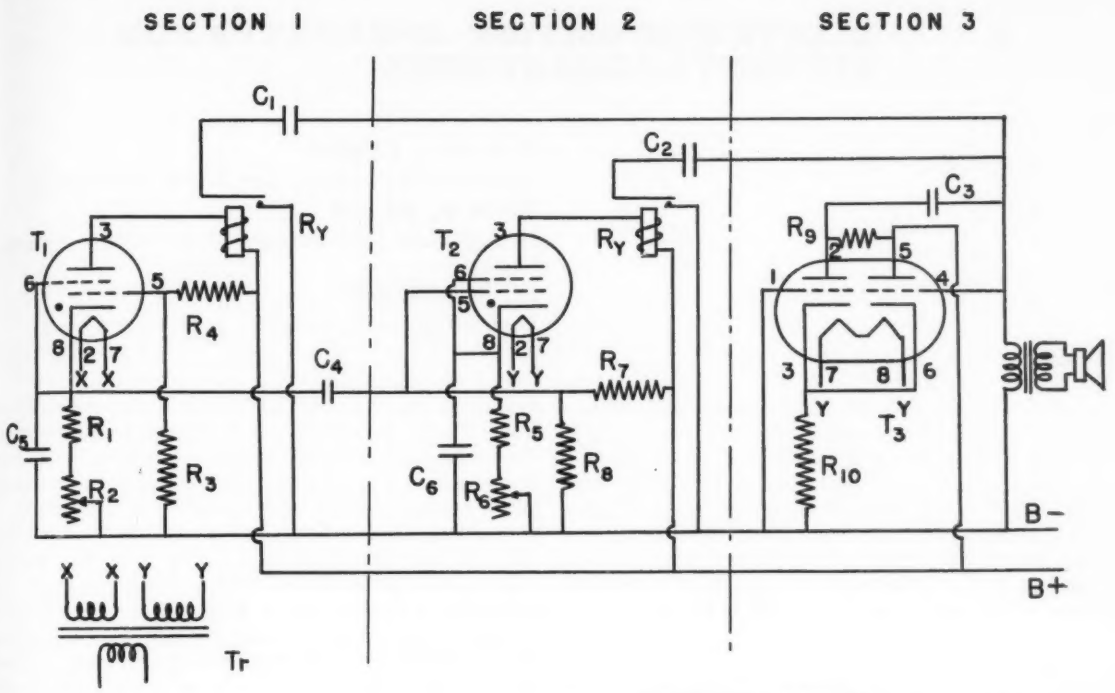
A STUDY of chemical kinetics ordinarily involves the measurement of the time elapsed before some evidence of a concentration change becomes apparent. While a watch or clock fitted with a sweep second hand is usually adequate in an elementary chemistry course where fast reactions are customarily employed, there is something to be gained by providing an audible time signal rather than a visual one, for the student can then concentrate on carefully mixing the reagents at time zero and on continuous observation for the change. The device to be described provides a tone impulse at a rate variable from 30 to 120 per minute and an additional characteristic signal for submultiples within a range of 5 to 15 per minute. Adjusted for normal use the apparatus emits a 400 cycle note every second and a 1000 cycle tone every tenth second.

The instrument consists of three distinct units independent enough to permit any to be omitted with no loss of function of the remainder. Sections 1 and 2 are multivibrator circuits with values of C and R selected to give the two different frequency ranges, while section 3 is a modification of an audio oscillator previously described.[1] If one merely requires a single signal at equal intervals section 2 can be deleted and the proper values of capacitance and resistance selected. Further simplification is possible, for section 3 can be eliminated if the noise level in the laboratory is low enough to permit the click made by the closing relay to be audible.

A power source capable of furnishing a maximum of 175 volts d.c. at 25 ma. is required in addition to the components called for by the circuit diagram. The d.-c. potential necessary for proper operation is not critical, for there is but little change in the beat rate when the voltage is reduced to 110 volts from the recommended 175 volts. There is, however, a noticeable drop in the output of the audio oscillator at lower plate potentials. It is important that each of the Type-2050 tubes be operated from a separate filament winding although the 6SN7GT can obtain its heater voltage from the source supplying T_2.

The beat rate is determined by the C-R combination between the cathode (pin 8) of the thyratron and ground. While the diagram specifies a 5-mfd. condenser, values of 2 mfd. and of 10 mfd. were also tried. A 2-mfd. capacitor passes about 2 ma. of plate current and requires a resistance of 0.4 megohm for a beat of 60 per minute, while the 10-mfd. condenser passes approximately 6 ma. and produces the same beat rate with a resistance of 0.1 megohm. One could, therefore, use any high-quality, oil-filled capacitor and obtain the same rate by suitable selection of the parallel resistance, but too low a value of capacitance may not permit the flow of sufficient plate current to energize the relay. Not

[1] MARION, ALEXANDER, P., J. CHEM. EDUC., **24**, 394 (1947).



C_1	0.01 mfd.
C_2	0.005 mfd.
C_3, C_4	0.1 mfd.
C_5, C_6	5 mfd., 400 volt, oil filled
R_1	75,000 ohms
R_2	0.25 megohm potentiometer
R_3, R_8	0.47 megohm
R_4, R_7	1 megohm
R_5	0.47 megohm + 0.27 megohm
R_6	2 megohm potentiometer
R_9	0.1 megohm
R_{10}	3,000 ohm
Ry	3 ma., normally open relay
T_1, T_2	Type 2050 tube
T_3	6 SN 7 GT
Tr	Filament transformer; 6.3 v. at 1.2 amp. (Y-Y), 6.3 v. at 0.6 amp. (X-X)

only should these capacitors have low leakage but they must be capable of withstanding a d.-c. potential of at least 150 volts. The values specified in the parts list allow an ample safety factor. The fixed resistors R_1 and R_3 are protective in nature.

The rather large value of the capacitor coupling the two multivibrators has an effect on the beat rate, for if T_1 is removed from its socket, section 2 has a range of from 3 to 8 per minute while its normal range with T_1 operative is from 5 to 15 per minute. However, with the value of capacitance (250 mmfd.) suggested in the literature[2] it was found that a critical adjustment of R_6 was necessary to synchronize the second thyratron on the desired submultiple, while experiment showed the larger value of coupling condenser easily "locks" the two circuits.

The relays used in the plate circuits of the thyratrons must be sensitive enough to respond to about 3 ma. The units actually employed are available at radio supply houses as war-surplus, differential relays containing dual 8000-ohm coils and bearing a part number of 803476. A separate relay was used for each tube, since attempts to use both sections of one relay gave unreliable results, apparently because of the interaction between the magnetic fields as the currents built up.

It should be pointed out that the tone developed by the audio oscillator depends upon the combination of inductance and capacitance between the grid (pin 4) of the 6SN7GT and ground. The primary of an output transformer is utilized for the inductance and, while a Jensen G8RS speaker and a matching transformer originally intended to couple a 6L6 to the voice coil were used because they were available, it is suggested that any transformer with a measured inductance of between four and seven henries be chosen and the proper tone be developed by an adjustment of the values of C_1 and C_2. Because of the time constant the submultiple signal commences as that produced by the master multivibrator but rapidly breaks into its own tone.

No audio amplification was incorporated in the circuit since the output proved sufficient for a group of 24 students in a reasonably quiet laboratory. Initially the tone generator was omitted with the hope that the clicking of the relay would provide an adequate signal, every submultiple being slightly louder because of the coincidence of both relays sounding. But while it functioned satisfactorily for use by one or two individuals, the signal strength was too weak for class use.

[2] TAYLOR, ALBERT H., *Radio and Television News*, **43**, 63 (Feb., 1950).

ELECTRODEPOSITION APPARATUS FOR STUDENT LABORATORIES

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THERE are good pedagogic reasons for including in elementary quantitative analysis at least one electrodeposition determination, generally that of copper in a copper-base alloy. It illustrates a different, yet important, type of separation preliminary to gravimetric measurement; it may be conveniently used to review (or initiate) the study of the basic relationships involved in electrochemical processes; it is an excellent introduction to the topic of oxidation and reduction.

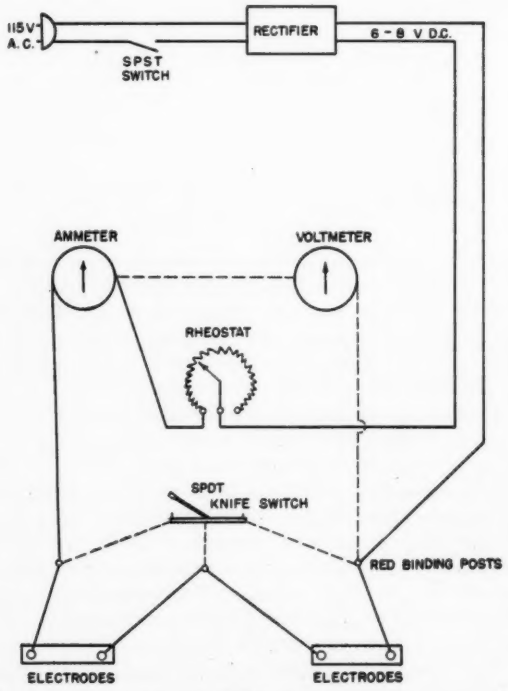

Figure 1. Simplified Schematic Diagram

One deterrent to a wider adoption of an electrodeposition determination in the elementary course has been the rather high price of the equipment required. The commercial product, though in general well designed and constructed, is not essential in the undergraduate laboratory. The simple apparatus described has given excellent service. It is easily constructed from readily available and inexpensive components, allowing a considerable number to be made available to the class; it is quite portable so that even the most petite feminine student has no difficulty in carrying it to her laboratory desk; it operates directly from the 115-volt a.-c. line; the open wiring panel has proved very successful in demonstrating to the student the simplicity of the equipment and the essential principles of the method. (Too frequently the less inquisitive student using a commercial piece of apparatus completes an experiment with only a very hazy idea of how the equipment functions.) To assist the student he is given a simplified schematic diagram (Figure 1).

The details of construction will be apparent from the photograph (Figure 2), the construction drawing (Figure 3), and the wiring diagram (Figure 4). The essential electrical parts are listed, together with a sug-

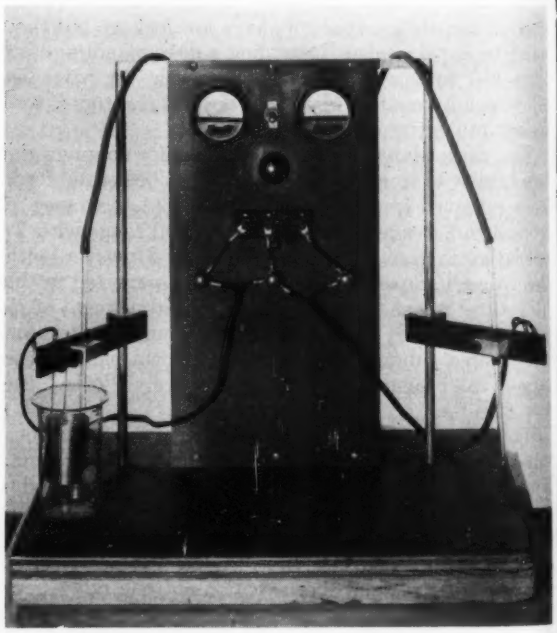

Figure 2. Student Electrodeposition Apparatus

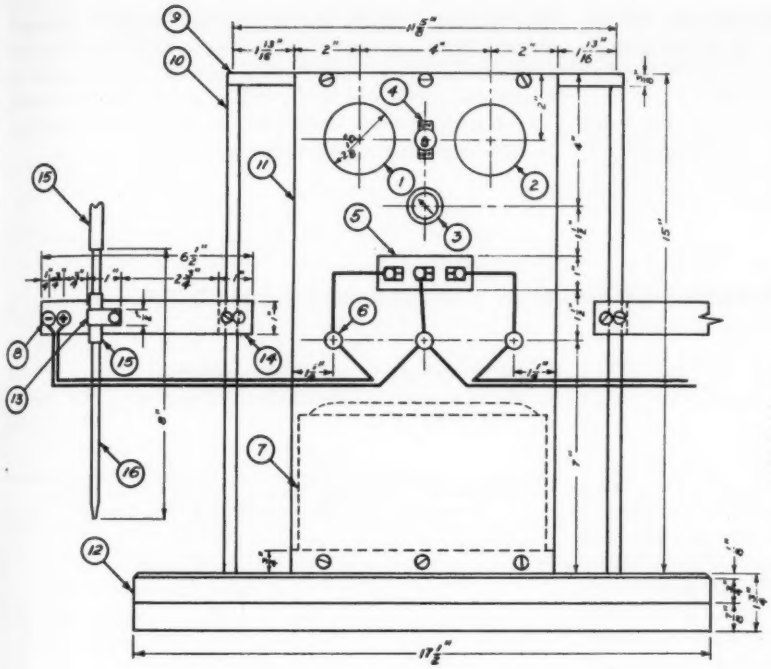

Figure 3. Constructional Details

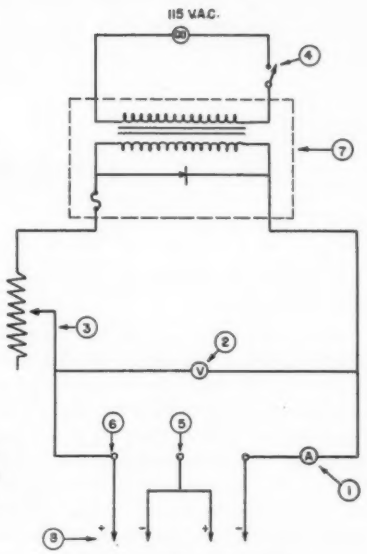

Figure 4. Wiring Diagram

gested source and the current price. The electrical parts cost about $17 while the constructional parts are usually available in the chemistry shop or can be purchased for a few dollars.

The base of the instrument is constructed of 1-inch wood, 5/8-in. plywood, and 1/8-in. hardboard (Masonite). The supporting metal framework is 3/8-in. brass, while the split sliding blocks which support the electrode arms and the air stirrers are of aluminum. The main panel and the side arms carrying the electrode terminals are of 1/4-in. Formica. It is obvious that other materials could be used. The side arms are fastened by two screws to the split block while the latter is fastened to the vertical rod by a screw passing through the split.

For simplicity, stirring of solutions is accomplished by bubbling air through a capillary into the solution. The line (rubber tubing) from the compressed air outlet is divided by a glass Y-tube into two lines (rubber tubing), which lead to each capillary. An adjustable screw clamp is inserted on one or both of the air lines to the capillaries to regulate and equalize the amount of stirring in each solution.

Several dozen of the assemblies have been in use in undergraduate laboratories for some years with excellent results and a minimum of maintenance. One unit is usually provided for each laboratory bench, which accommodates six students per section. By preparing the solutions beforehand, a student can readily complete a determination in duplicate during a three-hour period.

The instructions given the students for the operation of the apparatus are essentially those found in any standard textbook on elementary quantitative analysis plus the following details pertaining to the use of the present apparatus:

After the electrodes are in place, insert the air tube so that its tip is about 1/4-in. above the beaker bottom. Adjust the air current to give a rapid stream of bubbles

Code Numbers for Figures 3 and 4

1.	Ammeter	11.	1/8-in. Formica Panel
2.	Voltmeter	12.	5/8-in., 5-ply wood
3.	Rheostat	13.	Aluminum block, 1/2 × 3/4 × 1 in., drilled to receive glass tube
4.	Off-On switch		
5.	Knife switch		
6.	Binding posts, red	14.	Formica electrode side arm
7.	Battery charger		
8.	Binding posts, black	15.	Rubber sleeve, and tubing
9.	3/8-in. square brass	16.	8-mm. glass tubing, 8 in. long
10.	3/8-in. brass rod		

Electrical Components

Quantity Required	*Description*	*Catalogue No. Lafayette-Concord Radio Co.*	*Price*
1	Rheostat, 25-watt, 50-ohm	8N8980	1.76
1	Voltmeter, d. c., 0–8 volts	25N11055	1.27
1	Ammeter, d. c., 0–3 amp.	25N11092	1.27
1	Knife switch, SPDT	9N16831	0.59
1	Toggle switch, SPST	9N12836	0.29
1	Off-On switch plate	9N12818	0.03
1	Battery charger, 6AC4	21N15696	9.76
3	Insulated binding posts, red	13N13163	0.33
4	Eby binding post	13N13082	0.60
24 in.	Hook up wire (two colors)		

without splashing. Commence the electrolysis, adjusting the resistance so that a current of 0.6 to 0.8 amp. is obtained at a potential of about 2 to 3 volts (remember that the two solutions are connected in series).

To remove the electrodes, raise one set of electrodes slowly from the solution, with the current left on. When electrodeposition is complete, wash the exposed cathode with a stream of water from a wash bottle as the electrode is being raised. As soon as the cathode is above the surface of the solution, throw the short-out switch and continue deposition in the other beaker for an additional five or ten minutes.

A. C. S. LEADERS TRAINED IN LIBERAL ARTS COLLEGES

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SURVEYS on the origins of American scientists have shown that liberal arts colleges have a higher proportion of students continuing to the Ph.D. in the physical sciences than any other type of institution of higher learning.[1] Sixty-seven members of the National Academy of Sciences received their undergraduate training in liberal arts colleges.[2] The present study has been undertaken to determine what contributions these small colleges have made to leadership in the American Chemical Society.

Nine A. C. S. presidents since 1900 have been graduates of liberal arts colleges. The table lists the institutions which trained these leaders of American chemistry. The booklet, "Awards Administered by the American Chemical Society, 1952," contains the names of all recipients of the 11 prizes now given by the Society. The liberal arts colleges which trained the 18 recipients of these awards are listed in the table. Awards administered by local sections of the A. C. S. are not included. "The College Blue Book" has been used to ascertain the liberal arts status of the colleges. Institutions which grant the Ph.D. degree, or which have professional schools of engineering, medicine, and other applied sciences are not classed as liberal arts. "American Men of Science," and "Who's Who in America" have been consulted for the biographical data.

From this survey it can be concluded that one in five presidents of the A. C. S. during this century received his undergraduate training in a liberal arts college, and that about the same proportion applies to recipients of Society-administered awards.

Liberal Arts Colleges Which Have Trained A.C.S. Presidents and Recipients of A.C.S. Awards

Institution	*A.C.S. Presidents*	*Pure Chemistry Award*	*Borden Award*	*Eli Lilly Award*	*Fisher Award*	*Garvan Medal*	*Paul-Lewis Award*	*Priestley Medal*
Allegheny	..	..	..	..	1			
Amherst	..	1						
Battle Creek	..	1						
Butler	..	1						
Carleton	..	..	1					
Centenary C. for Women	..	..	..	..	..	1		
DePauw	..	..	..	2				
Dickinson	..	..	..	1				
Goucher	..	..	..	..	..	1		
Grinnell	2	..	..	..	..	..	..	1
Haverford	1	..	..	1				
Hobart	..	..	..	1				
Ill. Wesleyan	1							
Miami (Ohio)	1							
Okla. C. for Women	..	..	..	..	..	1		
Randolph-Macon	..	..	..	..	..	1		
Southwestern (Kan.)	..	1						
Transylvania	1							
Washington and Lee	1							
Wesleyan	..	..	..	..	..	..	1	
Westminster	1							
Williams	1	..	1					

[1] KNAPP, R. H., AND H. B. GOODRICH, *Science*, **113**, 543 (1951).
[2] SAMPEY, J. R., "Liberal arts colleges and the National Academy of Sciences," *Science*, **116**, 309 (1952).

CHEMICAL EDUCATION AWARD

WE WOULD like to remind our readers that June 1 is the deadline for nominations for the Scientific Apparatus Makers Award in Chemical Education. Details of the nomination procedure may be found on page 167 of our March, 1951, issue.

THE HYDROGEN ION CONCENTRATION OF WEAK MONOBASIC ACIDS AND THEIR SALTS[1]

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ACIDS ONLY

Let HA represent a monobasic acid which does not ionize completely when dissolved in water. Then in a water solution the following equilibria must be satisfied:

$$\frac{[H^+][A^-]}{[HA]} = K_a \qquad (1)$$

which may be rearranged to

$$[HA] = \frac{[H^+][A^-]}{K_a}$$

and

$$[H^+][OH^-] = K_w \qquad (2)$$

The acid must exist in either the ionized or the molecular form and the sum of these concentrations is the total concentration of the acid, so that

$$[HA] + [A^-] = C_a \qquad (3)$$

Because the solution is electrically neutral, the number of positive and negative charges must be equal, and

$$[H^+] = [A^-] + [OH^-] \qquad (4)$$

Substitution of (1) in (3) gives

$$[A^-] = \frac{C_a K_a}{[H^+] + K_a}$$

Substitution of (2) in (4) gives

$$[A^-] = \frac{[H^+]^2 - K_w}{[H^+]}$$

Equating and rearranging gives

$$[H^+]^3 + K_a[H^+]^2 - C_aK_a[H^+] - K_w[H^+] - K_aK_w = 0 \qquad (5)$$

which is the relationship between $[H^+]$ and C_a for any monobasic acid.

The relative importance of the terms in equation (5) depends upon the values of K_a and C_a as may be seen in Table 1. Table 1 also contains calculated values for the pH of a number of weak acid solutions. The numbers in the rows opposite the individual terms are approximate values for the negative logarithms of each term, *i. e.*, the px values, where $px = -\log x$. All terms with relatively large px values may be dropped without diminishing the accuracy of the calculation. The following equations may be obtained from (5) by considering only the terms specified.

Terms	*Equation*
1, 3	$[H^+] = \sqrt{C_aK_a}$
1, 2, 3	$[H^+] = \sqrt{\left(\frac{K_a}{2}\right)^2 + C_aK_a} - \frac{K_a}{2}$
1, 3, 4	$[H^+] = \sqrt{C_aK_a + K_w}$

[1] Presented at the 122nd Meeting of the American Chemical Society, Atlantic City, September, 1952.

TABLE 1
Calculated pH Values of Weak Acid Solutions

pK_a	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*
				$C_a = 0.1\ M$						
1 p$[H^+]^3$	3.6	4.7	6.1	7.5	9.0	10.5	12	13.5	15	16.5
2 p$K_a[H^+]^2$	3.4	5.1	7.0	9.0	11.0	13	15	17	19	21
3 p$C_aK_a[H^+]$	3.2	4.6	6.0	7.5	9.0	10.5	12	13.5	15	16.5
4 p$K_w[H^+]$	15	15.6	16	16.5	17	17.5	18	18.5	19	19.5
5 pK_aK_w	15	16	17	18	19	20	21	22	23	24
pH 1, 3	1.00	1.50	2.00	2.50	3.00	3.50	4.00	4.50	5.00	5.50
pH 1, 2, 3	1.21	1.57	2.02	2.51	3.00	...	...	...	...	...
				$C_a = 0.001\ M$						
1 p$[H^+]^3$	9.0	9.1	9.6	10.7	12	13.5	15	16.5	18	19.5
2 p$K_a[H^+]^2$	7.0	8.0	9.4	11.1	13	15.0	17	19	21	23
3 p$C_aK_a[H^+]$	7.0	8.0	9.2	10.6	12	13.5	15	16.5	18	19.5
4 p$K_w[H^+]$	17	17	17	17.6	18	18.5	19	19.5	20	20.5
5 pK_aK_w	15	16	17	18	19	20	21	22	23	24
pH 1, 3	2.00	2.50	3.00	3.50	4.00	4.50	5.00	5.50	6.00	6.50
pH 1, 2, 3	3.00	3.04	3.21	3.57	4.02	4.51	5.00	...	...	...
pH 1, 3, 4	...	...	...	...	...	...	...	...	...	6.48

SALTS ONLY

Salts of Weak Acids and Strong Bases

Let BA represent the salt of a strong base and a weak monobasic acid. In a water solution equations (1) and (2) still apply. Let C_s be the effective salt concentration, then

$$C_s = [B^+] \quad (6)$$

and

$$C_s = [HA] + [A^-] \quad (7)$$

Because of electrical neutrality

$$[B^+] + [H^+] = [A^-] + [OH^-] \quad (8)$$

Substituting (1) in (7), (2) in (8), and solving as before gives

$$[H^+]^3 + K_a[H^+]^2 + C_s[H^+]^2 - K_w[H^+] - K_aK_w = 0 \quad (9)$$

TABLE 2

Calculated pH Values of Solutions of Salts of Weak Acids and Strong Bases

pK_a	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*
					$C_s = 0.1\ M$					
1 $p[H^+]^3$	21.5	22.5	24	25.5	27	28.5	30	31.5	33	34.5
2 $pK_a[H^+]^2$	15.3	17	19	21	23	25	27	29	31	33
3 $pC_s[H^+]^2$	15.3	16	17	18	19	20	21	22	23	24
4 $pK_w[H^+]$	21	21.5	22	22.5	23	23.5	24	24.5	25	25.5
5 pK_aK_w	15	16	17	18	19	20	21	22	23	24
pH 3, 5	7.00	7.50	8.00	8.50	9.00	9.50	10.00	10.50	11.00	11.50
pH 2, 3, 5	7.15	7.52	8.00	...	...	...	...	...	...	...
					$C_s = 0.001\ M$					
1 $p[H^+]^3$	21	21	21.5	22.5	24	25.5	27	28.5	30	31.3
2 $pK_a[H^+]^2$	15	16	17.3	19	21	23	25	27	29	30.9
3 $pC_s[H^+]^2$	17	17	17.3	18	19	20	21	22	23	23.9
4 $pK_w[H^+]$	21	21	21	21.5	22	22.5	23	23.5	24	24.4
5 pK_aK_w	15	16	17	18	19	20	21	22	23	24
pH 3, 5	6.00	6.50	7.00	7.50	8.00	8.50	9.00	9.50	10.00	10.50
pH 2, 3, 5	7.00	7.02	7.15	7.52	8.00	...	...	...	...	...
pH 3, 4, 5	...	...	...	...	...	...	...	...	9.98	10.43

TABLE 3

Hydrogen Ion Concentrations of Various Ammonium Salt Solutions

(Calculated from Equation 11)

Term	*HIO_3*	*$Cl_2C_2H_2O_2$*	*$ClC_2H_3O_2$*	*H_2CO_2*	*$C_2H_4O_2$*	*$C_5H_4O_2N_4$*	*$C_3H_3O_3N_3$*	*$HClO$*	*HCN*
					0.1 *M*				
1 $K_b[H^+]^4$	26	26	29	30	33	35	39	41	42
2 $C_sK_b[H^+]^3$	21	22	24	25	27	29	30	31	33
3 $K_aK_b[H^+]^3$	21	22	26	28	31	33	35	37	42
4 $K_w[H^+]^3$	30	30	32	33	35	37	38	39	42
5 $K_aK_w[H^+]^2$	25	26	29	31	33	35	37	39	42
6 $K_bK_w[H^+]^2$	29	29	31	32	33	34	35	36	37
7 $C_sK_aK_w[H^+]$	21	22	25	25	27	28	30	31	33
8 $K_aK_bK_w[H^+]$	25	25	28	29	31	32	34	34	37
9 $[K_w]^2[H^+]$	33	33	34	34	35	36	36	36	37
10 $K_a[K_w]^2$	29	29	31	32	33	34	35	35	37
pK_a	0.72	1.30	2.80	3.68	4.75	5.82	6.70	7.43	9.14
$[H^+]$ 2, 7	1.0×10^{-5}	5.3×10^{-6}	9.4×10^{-7}	3.4×10^{-7}	1.0×10^{-7}	2.9×10^{-8}	1.1×10^{-8}	4.5×10^{-9}	6.3×10^{-10}
$[H^+]$ 2, 3, 7	6.0×10^{-6}	4.3×10^{-6}	9.3×10^{-7}	3.4×10^{-7}	...	...	...	...	...
pH	5.22	5.37	6.03	6.47	7.00	7.54	7.98	8.34	9.20
					0.001 *M*				
1 as above	29	29	30	30	33	35	39	41	42
2 as above	26	26	26	27	29	31	32	33	35
3 as above	24	25	26	28	31	33	35	37	42
4 as above	32	32	33	34	35	37	38	39	42
5 as above	27	28	29	31	33	35	37	39	42
6 as above	31	31	31	32	33	34	35	36	37
7 as above	24	25	26	27	29	30	32	33	35
8 as above	26	27	27	29	31	32	34	34	37
9 as above	34	34	34	35	35	36	36	36	37
10 as above	29	29	31	32	33	34	35	35	37
$[H^+]$ 2, 7	1.0×10^{-5}	5.3×10^{-6}	9.4×10^{-7}	3.4×10^{-7}	1.0×10^{-7}	2.9×10^{-8}	1.1×10^{-8}	4.5×10^{-9}	6.3×10^{-10}
$[H^+]$ 3, 7	7.5×10^{-7}	7.5×10^{-7}	7.5×10^{-7}	...	...	...	...	...	...
$[H^+]$ 2, 3, 7	7.4×10^{-7}	7.4×10^{-7}	5.8×10^{-7}	3.1×10^{-7}	...	...	...	...	...
$[H^+]$ 2, 7, 8	...	...	...	...	...	2.9×10^{-8}	1.1×10^{-8}	4.6×10^{-9}	...
$[H^+]$ 2, 3, 7, 8	7.5×10^{-7}	7.5×10^{-7}	5.9×10^{-7}	3.1×10^{-7}	1.0×10^{-7}	2.9×10^{-8}	...	...	...
$[H^+]$ 2, 6, 7, 8, 9, 10	...	...	...	...	...	...	...	...	6.5×10^{-10}
pH	6.12	6.12	6.23	6.51	7.00	7.54	7.97	8.34	9.19

This equation differs from (5) in the third term only. However, the relative importance of the terms is different as may be seen in Table 2. The following equations may be obtained from (9) by considering only the terms specified:

Terms	*Equation*
3, 5	$[H^+] = \sqrt{\frac{K_a K_w}{C_s}}$
2, 3, 5	$[H^+] = \sqrt{\frac{K_a K_w}{C_s + K_a}}$
3, 4, 5	$[H^+] = \sqrt{\left(\frac{K_w}{2C_s}\right)^2 + \frac{K_a K_w}{C_s}} + \frac{K_w}{2C_s}$

Table 2 contains px and pH values, calculated from the appropriate equations for a number of representative salts.

Salts of Weak Acids and Weak Bases

In addition to the equilibrium equations (1) and (2),

$$\frac{[B^+][OH^-]}{[BOH]} = K_b \qquad (10)$$

is needed. Let C_s be the effective salt concentration; then

$$C_s = [B^+] + [BOH]$$

and

$$C_s = [A^-] + [HA]$$

Equation (8) still applies. By substitution and simplification the following equation may be obtained:

$$K_b[H^+]^4 + C_s K_b[H^+]^3 + K_a K_b[H^+]^3 + K_w[H^+]^3 + K_a K_w[H^+]^2 - K_b K_w[H^+]^2 - C_s K_a K_w[H^+] - K_a K_b K_w[H^+] - [K_w]^2[H^+] - K_a[K_w]^2 = 0 \qquad (11)$$

As a rule only terms number 2, 3, 7, and 8 are applicable. In very dilute solutions of the weakest acids terms 9 and 10 become significant.

The following approximate equations may be obtained by consideration of the indicated terms.

TABLE 4

Calculated pH Values of Solutions of Weak Acids Plus Their Salts of Strong Bases

pK_a	*1*	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*
				$C_s = C_a = 0.1\ M$						
1 p$[H^+]^3$	4.1	6.2	9.0	12.0	15	18	21	24	27	30
2 p$K_a[H^+]^2$	3.8	6.1	9.0	12	15	18	21	24	27	30
3 p$C_s[H^+]^2$	3.8	6.1	7.0	9	11	13	15	17	19	21
4 p$C_a K_a[H^+]$	3.4	5.1	7.0	9	11	13	15	17	19	21
5 p$K_w[H^+]$	15.4	16	17	18	19	20	21	22	23	24
6 p$K_a K_w$	15	16	17	18	19	20	21	22	23	24
pH 3, 4	1.00	2.00	3.00	4.00	5.00	6.00	7.00	8.00	9.00	10.00
pH 1, 2, 3, 4	1.38	2.07	3.00	...	...	...	...	...	...	...
				$C_s = C_a = 0.001\ M$						
1 p$[H^+]^3$	9.0	9.0	10.2	12.2	15	18	21	24	27	30
2 p$K_a[H^+]^2$	7.0	8.0	9.7	12.1	15	18	21	24	27	30
3 p$C_s[H^+]^2$	9.0	9.0	9.7	11.1	13	15	17	19	21	23
4 p$C_a K_a[H^+]$	7.0	8.0	9.4	11.1	13	15	17	19	21	23
5 p$K_w[H^+]$	17	17	17.4	18	19	20	21	22	23	24
6 p$K_a K_w$	15	16	17	18	19	20	21	22	23	24
pH 3, 4	1.00	2.00	3.00	4.00	5.00	6.00	7.00	8.00	9.00	10.00
pH 2, 4	3.00	3.00	3.00	...	...	...	...	...	...	...
pH 1, 2, 3, 4	3.00	3.07	3.38	4.07	5.00	...	...	...	...	...
pH 3, 4, 5, 6	...	...	...	...	...	...	...	...	...	9.93
				$C_s = 0.001\ M$	$C_a = 0.1\ M$					
1 p$[H^+]^3$	3.6	4.7	6.1	7.7	9.6	12	15	18	21	24
2 p$K_a[H^+]^2$	3.4	5.1	7.1	9.1	11.4	14	17	20	23	26
3 p$C_s[H^+]^2$	5.4	6 1	7.1	8.1	9.4	11	13	15	17	19
4 p$C_a K_a[H^+]$	3.2	3.6	6.0	6.6	9.2	11	13	15	17	19
5 p$K_w[H^+]$	15	16	16	17	17	18	19	20	21	22
6 p$K_a K_w$	15	16	17	18	19	20	21	22	23	24
pH 3, 4	−1	0	1	2	3	4	5	6	7	8
pH 1, 4	1	1.5	2.0	2.5	3	...	...	...	...	...
pH 1, 2, 4	1.21	1.57	2.02	2.51	...	...	...	...	...	...
pH 1, 3, 4	...	...	...	2.57	3.21	4.04	5.00	...	...	...
pH 1, 2, 3, 4	...	1.57	2.04	2.57	...	...	...	...	...	...
				$C_s = 0.1\ M$	$C_a = 0.001\ M$					
1 p$[H^+]^3$	9.9	12.1	15	18	21	24	27	30	32	33
2 p$K_a[H^+]^2$	7.6	10.1	13	16	19	22	25	28	31	32
3 p$C_s[H^+]^2$	7.6	9.1	11	13	15	17	19	21	23	23
4 p$C_a K_a[H^+]$	7.3	9.0	11	13	15	17	19	21	23	24
5 p$K_w[H^+]$	17	18	19	20	21	22	23	24	25	26
6 p$K_a K_w$	15	16	17	18	19	20	21	22	23	24
pH 3, 4	3	4	5	6	7	8	9	10	11	12
pH 2, 3, 4	3.30	4.04	5.00	...	...	...	...	...	...	...
pH 3, 4, 6	...	...	...	...	...	...	9.00	9.96	10.79	10.97

Terms	*Equation*
2, 7	$[H^+] = \sqrt{\frac{K_aK_w}{K_b}}$
3, 7	$[H^+] = \sqrt{\frac{C_aK_w}{K_b}}$
2, 3, 7	$[H^+] = \sqrt{\frac{C_aK_aK_w}{C_aK_b + K_aK_b}}$
2, 7, 8	$[H^+] = \sqrt{\frac{C_aK_aK_w + K_aK_bK_w}{C_aK_b}}$
2, 3, 7, 8	$[H^+] = \sqrt{\frac{C_aK_aK_w + K_aK_bK_w}{C_aK_b + K_aK_b}}$

Table 3 contains px and pH values calculated for a number of representative ammonium salts.

ACIDS AND SALTS TOGETHER

Salts of Strong Bases and Weak Acids

Equations (1), (2), (6), and (8) still apply. In addition,

$$C_a + C_s = [HA] + [A^-] \qquad (12)$$

From these relationships the following equation may be derived:

$$[H^+]^3 + K_a[H^+]^2 + C_s[H^+]^2 - C_aK_a[H^+] - K_w[H^+] - K_aK_w = 0 \qquad (13)$$

When C_s is zero this simplifies to (5). When C_a is zero it reduces to (9). The fifth term in (13) is rarely significant, but each of the others becomes important under certain conditions. Table 4 lists calculated values of px and pH. In the construction of this table the following approximate equations derived from (13) were used:

Terms	*Equation*
3, 4	$[H^+] = \frac{C_aK_a}{C_s}$
1, 4	$[H^+] = \sqrt{C_aK_a}$
1, 3, 4	$[H^+] = \sqrt{\left(\frac{C_s}{2}\right)^2 + C_aK_a} - \frac{C_s}{2}$
1, 2, 4	$[H^+] = \sqrt{\left(\frac{K_a}{2}\right)^2 + C_aK_a} - \frac{K_a}{2}$
2, 3, 4	$[H^+] = \frac{C_aK_a}{K_a + C_s}$
3, 4, 6	$[H^+] = \sqrt{\left(\frac{C_aK_a}{2C_s}\right)^2 + \frac{K_aK_w}{C_s}} + \frac{C_aK_a}{2C_s}$
1, 2, 3, 4	$[H^+] = \sqrt{\left(\frac{C_s + K_a}{2}\right)^2 + C_aK_a} - \frac{C_s + K_a}{2}$
3, 4, 5, 6	$[H^+] = \sqrt{\left(\frac{C_aK_a + K_w}{2C_s}\right)^2 + \frac{K_aK_w}{C_s}} + \frac{C_aK_a + K_a}{2C_s}$

PROPERTIES AND THE PERIODIC TABLE

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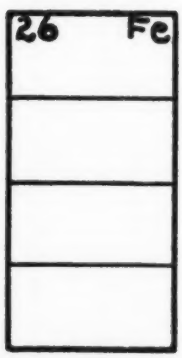
A NUMBER of investigators have presented certain properties of elements by use of the long form of the periodic table.[1,2] This device is an excellent method for showing the tendencies by family and by group of common physical and chemical properties.

One drawback to this fundamental presentation has been the lack of suitable forms or charts for the recording of such data. Another factor that has retarded the use of such presentation has been the disregard by many investigators of the fact that compounds of the elements exhibit similar characteristic trends by family and by group. The historical prediction of properties of elements by the properties of surrounding elements before the element itself was isolated may also be used in like sense for compounds of the elements.

Fundamental work concerning metallic formates in this laboratory led to an investigation of chemical and physical properties of all of the formates reported in the literature to this time. Tabulation of these data was best done in periodic form; experience gained in the construction of this chart will be of value to other investigators. The chart consisted of the long form of the periodic table, with two rows below for the lanthanide and actinide series. The space for each element was a rectangle 1 inch wide and 2 inches deep. These dimensions were chosen in order to provide adequate space for entering data, at the same time keeping the size of the chart within reasonable limits. The symbol and atomic number of each element were placed in the proper rectangle in as fine print as possible, the remaining space being free for additional data (see the figure).



One Element-Rectangle of the Chart, Actual Size

The rectangles for recording data in the chart were ruled off in several ways, depending on the type of data collected. For data concerning a single property the space was not ruled further; for multiple data such as

[1] HAZLEHURST, T. H., AND F. J. FORNOFF, J. CHEM. EDUC., 20, 77–9 (1943).
[2] KLINGENBERG, J., AND L. SPRINGMAN, *ibid.*, 29, 81 (1952).

melting point, boiling point, solubility, and the existence of hydrates, the spaces were ruled into four horizontal sections as indicated in the figure. Adequate space is present on the chart, especially between beryllium and boron and magnesium and aluminum, for explanatory symbols and codes.

Copies of the various types of this presentation can be made quite cheaply by use of a reproduction method of the Ozalid type, or can be printed in pads for greater use. Advanced undergraduate and graduate instruction, as well as fundamental research, can be greatly enhanced by the use of such charts.

A CONTINUOUS LIQUID-LIQUID EXTRACTOR FOR ORGANIC SOLVENTS DENSER THAN WATER

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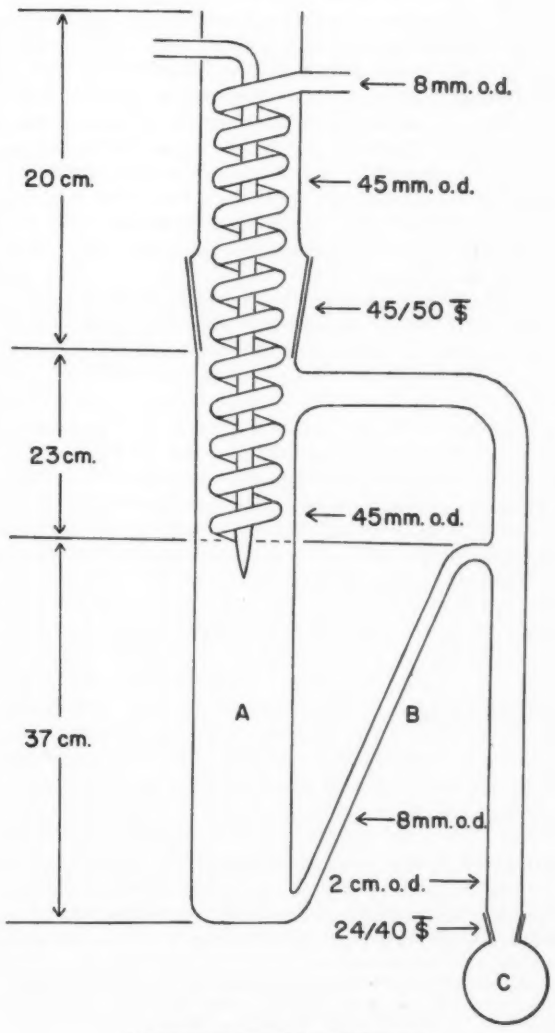
EXTRACTION of aqueous solutions with denser organic solvents has been accomplished conveniently, with good temperature control, using the simple apparatus shown.

The extraction chamber *A*, and the outer wall of the coil condenser, were constructed from 45-mm. o.d. Pyrex tubing; to permit temperature control, the condenser coil extended into the aqueous solution to a point about even with the uppermost level of the solvent return line *B*. The length of chamber *A* and the height of the top of the return line *B* are critical; the dimensions given in the diagram have been found satisfactory for methylene chloride extraction of dilute aqueous solutions up to about 350 ml. in volume.

To carry out an extraction, about 100 ml. of the organic solvent is placed in chamber *A*, and then the aqueous solution is introduced. The condenser unit is placed in position, and the organic solvent in flask *C* boiled vigorously. Drops of organic liquid up to 1 cm. in diameter begin to descend through the aqueous solution. If small volumes of solution are to be extracted, the required liquid level in chamber *A* may be maintained, with a reasonable volume of organic solvent, by introducing 30-cm. lengths of glass rod.

The efficiency of the apparatus was determined by extraction of 1 *N* acetic acid with methylene chloride and titration of aliquots of the two phases in chamber *A*. Acetic acid in the organic phase was found to attain 76 per cent of its equilibrium concentration as determined with use of a separatory funnel. This rate of extraction is comparable with that secured in the more complicated and less versatile apparatus used by Hershberg and Wolfe.[1]

In practical use, it has been found that the apparatus will handle systems which give very troublesome emulsions when extraction is attempted with a separatory funnel, or with ether in the usual continuous-type apparatus. A length of wire with a loop at its lower end, inserted through the top of the condenser and manipulated at the interface in chamber *A*, is useful in controlling emulsions.



A Continuous Liquid-Liquid Extractor

[1] *J. Biol. Chem.*, **141**, 215 (1941).

Report of the

NEW ENGLAND ASSOCIATION OF CHEMISTRY TEACHERS

RECENT RESEARCH ON SUGAR MAPLES[1]

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University of Vermont, Burlington, Vermont

THE maple industry is an old one in New England, and through the years a considerable amount of work has been done on the chemistry of maple sap and maple syrup. Much of this work was done by C. H. Jones, our station chemist, who addressed this Association here in Burlington in 1935. Recently a group of us in the Botany Department—Dr. Taylor, Dr. Sproston, myself, and others—have become much interested in maple. Rather than give you a detailed account of the research projects we are engaged in, I shall try to point out some of the areas where research should yield important fundamental information and at the same time benefit the industry.

Most of the commercially important substances secured from woody plants such as resins, gums, alkaloids, appear to be by-products of the plants' metabolism. The function of these substances in the plan is not well understood. In contrast to these, maple sap is a solution of compounds which occur in the conducting elements in the plants and are used by the growing buds and the cambium to produce new growth. It may surprise you that even now we do not have a complete analysis of this sap. It is a dilute solution having, on the average, about 3 per cent solids and a pH of about 7. Of these solids approximately 95 per cent is sucrose, 1 per cent or less reducing sugars, 1.5 per cent ash, 1.5 per cent malic acid, and about 0.2 per cent protein as determined by the Kjeldahl method. The ash contains calcium, potassium, phosphorus, sodium, magnesium, manganese, silicon, sulfur, chlorine. There are also present, in addition to the malic acid, formic, acetic, citric, fumaric, and succinic acids. Recently the U. S. Eastern Regional Research Laboratory has been working over these organic acids and finding some additional ones. Dr. Sproston and his group here were interested in the nitrogen fraction, and they found a protein and a polypeptide. Most interesting of all was the fact that no free amino acids were present. Another one of our group, Miss Greene, is at the moment chromatographing sugars to see whether there are some additional sugars in minute amounts which we have not so far known about. If this should prove to be the case, they will be very interesting from a physiological point of view.

Maple flavor or flavors found in maple syrup and maple sugars are of considerable interest. Maple sap has little or no maple flavor. We have been processing fresh sap through a Strumia apparatus and reduced it to a freeze-dried solid material. The maple flavor develops on heating at atmospheric pressure. We do not know what this flavor is. Nelson has isolated an unstable phenolic substance which is similar in its odor and properties to vanillin. Snell, after repeated fractionations from a chloroform extract of syrup (1939), found a reddish viscous oil which developed a maple odor. Recently a synthetic maple flavor has been produced as the result of a browning reaction between alpha amino butyric acid and glucose, but the complete story on the parent compounds and the development of the flavor isn't known.

Those of you who have had experience in the food industry know how difficult it is to characterize some flavors. Two years ago one of the big blenders was interested in setting up taste panels and wanted known samples to use for references. One of the group came up here and we went about this area, talking with people who had had a good deal of experience with maple syrup. We went to one man who is a successful packer of pure maple. Unfortunately he was in bed with the grippe. His wife is a prominent member of this firm, and we told her our story, saying that we were trying to get a verbal description of the flavor characteristics of a good A or No. 1 syrup. She asked me where my home was and I had to confess that I had come originally from Connecticut, and the chap with me confessed that he was from Maine. The lady snorted somewhat and said, "Well, if you had been native born Vermonters you wouldn't have asked me this question, and since you are not, it is not possible for me to describe it to you."

As botanists we are interested in the maple tree. It is a wild plant, and fortunately our New England maples show many of the characteristics of a hybrid population.

[1] Based upon an address presented at the Fourteenth Summer Conference of the New England Association of Chemistry Teachers, University of Vermont, Burlington, Vermont, August 18, 1952.

One would expect to find variations in the three characteristics which are important to us, and which would be desirable in developing an improved stock for orchard planting. These characteristics are high sugar content, abundant flow, and rapid growth. Dr. Taylor has been studying the first of these, the variation of sugar content, over nine seasons and on several thousand trees. He found it desirable to make his determinations directly at the tree when the sap was flowing freely. Using hand refractometers it is possible to test about 100 trees an hour, and with several workers an entire sugar bush can be tested on any particular day. This is very important because he found that the sugar content of individual trees varies not only from season to season, but within the same season. The important point comes out, however, that a tree with high sugar content, although it may vary from time to time, maintains its position relative to the other trees in the population. We have found a very few trees with a sugar content much higher than average so that, from the point of view of this variable at least, we can expect something like a 100 per cent increase over the average figure of the sugar content of trees in the state.

The mechanism of sap flow in the spring is a problem very intriguing to botanists and still not entirely understood. Some of the earliest work was done in the '80's when it was shown that very considerable pressures occur in the stems of some plants, and that these pressures are responsible for the flow. Mr. Jones, about 1900, did a considerable amount of work in which he demonstrated that a physical explanation such as liquid expansion, or gas expansion or expansion of the wood could not account for the magnitudes found. The flow is clearly the response to a temperature rise, and it does not result from a movement from the roots as it is popularly pictured. We have done many experiments under conditions of controlled temperature using small sections of maple stems. Under these conditions a flow quite comparable in amount to that found in nature occurs. If the ends of these sections of stem are allowed to absorb water or a solution of sucrose, repeated flows can be produced. The technique is simple. The stems are cooled, frozen, and then warmed, the amount of flow measured and the rate observed. We have found that one can substitute the vessel sap from one tree for the vessel sap in another and get apparently normal flow. We can also wash out the vessel sap and put in a synthetic sap of sucrose with its concentration isosmotic with the sap in the plant and get a normal flow. If sugars other than sucrose are tried, disaccharides such as maltose and lactose cause a flow, also the trisaccharide, raffinose. If instead of using a sugar, distilled water is perfused through the stem after freezing and thawing, instead of a flow an absorption of water occurs. If hexose sugars are used, the physiological response is similar to that with distilled water—namely, they absorb. It is apparent that here we are not dealing with a simple osmotic system. If we were, one would suspect that we could substitute one chemical compound for another if the osmotic concentrations were the same, and membrane penetration did not occur. That is not the case. Depending upon the sugar, the mechanism either works well or does not work at all. Another reason that it does not seem to be a simple osmotic system is the fact that the rates and amounts of flow are not functions of the concentration of osmotically active substances in the sap. Osmosis may very well play a part, but if it does, it is one step in a multiple-factor mechanism.

There are one or two comments about the mechanism that can be made with certainty. If a flow occurs, it occurs on a rising temperature, and a falling temperature always causes a reduced rate, and frequently the flow stops. At this point you are probably thinking, "The temperature of *what?*" At our experimental maple farm in Underhill, Vermont, we have, for three seasons, kept accurate records of temperatures of the bark, of the wood, and of the twigs, using thermocouples and potentiometer recorders. At the same time we have been recording the rates and amounts of flow. From these observations it has become clear that the wood temperatures (and it is from the wood that this flow occurs) do not change very much. Rather the bark temperatures and the twig temperatures are the ones that show a marked diurnal fluctuation with which the flow rate changes correlate the best. It became evident very soon from these observations that a freezing temperature is not necessary. There is nothing mysterious about the effect of 32°F. or 0°C. on the flow mechanism. We have records of repeated flows in which neither the twigs, bark, nor wood got down to 0°C. It is essential, however, that there be a period of cooling before the next flow period. From that point of view, our diurnal cycle of 24 hours is not a particularly good one for maple flow. A 36- or 48-hour day would be very much better. Wood and bark are good insulators, and the few hours between sundown and sunrise are not sufficient to cool the wood very much. I think this explains why in the middle of the season the farmers say, "If we could only have a good wet snow that sticks to the trees. . . ." They mean that they would like to have a night and a day and another night of cooling temperatures, after which, with the normal daytime temperatures in the 50's, a temperature change of some 20°F. could be expected. That is sufficient to produce a good flow.

We are finding that just as in the case of sugar concentration so also there are individual differences in the flow-rate pattern. We have sufficient data to know that these two persist from season to season. This gives us considerable courage to go on with this sort of work and we hope ultimately to have available a quantity of pedigreed trees, with both superior sugar content and flow characteristics.

As you all know from current popular scientific literature, there is a good deal of work done with tree breeding. Our native maples grow rather slowly, at least more slowly than we would like. There is a maple in the Columbia River Valley which grows very rapidly. I have had them grow as much as five feet tall the first

year from seed. Unfortunately these plants are not hardy in Vermont and cannot take our rigorous winters. They do, however, flow maple sap and I have thought that hybridization, if possible, would be interesting. Consequently, using pollen from this western maple, we have been successful in getting a few hybrid seeds and are watching these new maples with great interest.

There is one question you might very well ask. "Does tapping injure the tree?" Tapping *per se* doesn't injure it. It isn't possible to take out enough sugar to injure or starve the tree unless one puts in some dozen or two spouts. However, the injury that results from tapping makes a wound which is easily infected and many of our good trees are going out of production as a result of decay from this injury. Dr. Sproston's group has isolated the disease-producing organisms, and we now know a good deal about it and are in the process of developing what we hope will be effective control measures.

I think perhaps this brief discussion will suffice to indicate that in the biology, physiology, biochemistry as well as pathology of our native trees there are interesting problems and important answers awaiting a solution in nearly any direction one may turn.

To the Editor:

For some time I have been using a somewhat different method of preparing punched cards for indexing information than that suggested by Dr. George R. Thomas in the August, 1952, issue of THIS JOURNAL.

I have found it convenient to prepare my punched cards by use of a punch of the type used in preparing papers for plastic binders similar to the spiral binder. These punches make a series of rectangular holes 7 × 3 mm. in size spaced 7 mm. apart, and will accommodate cards up to about 2 ft. on the side. By utilizing all four edges of the card I have 44 holes on a standard 5- × 8-in. file card.

In situations where a punch of this type is available I believe the preparation of cards is simpler than that described, and that the relatively long horizontal hole makes alignment of holes easier than the small round hole usually used. I find a microspatula handy for sorting.

We are using these cards for our master file of test items in addition to indexing information of other types.

RALPH E. CULLMANN

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BELLINGHAM, WASHINGTON

To the Editor:

D. F. Swinehart, in "More on oxidation numbers" [J. CHEM. EDUC., **29**, 284 (1952)], invites at least three questions. To his categorical statement "it is simply not necessary ever to ask this question" (what is the oxidation number of the element), the following query may be made: How did he decide that what he referred to as "oxidation half reaction" is oxidation, not reduction, or something else?

His starting with H in NH_4CNS seems arbitrary. How does one know what to begin with? With the other half reaction, for example?

He calls the ion-electron method "intellectually honest." How does he propose to explain:

$$2KMnO_4 + 16HCl \rightarrow 2MnCl_2 + 5Cl_2 + 8H_2O + 2KCl$$

Regarding your editorial of December, 1950, I would suggest the name *reaction.* The word connotes chemical interaction, and can be legitimately used whether the "equation" is complete, incomplete, balanced, or unbalanced. Thus, one might say: Complete and balance the following *reaction:*

$$HCl + Zn \rightarrow$$

Or, balance the following completed reaction:

$$HCl + Zn \rightarrow H_2 + ZnCl_2$$

Or, complete and balance the following incomplete reaction:

$$KMnO_4 + HCl \rightarrow MnCl_2 + Cl_2 + \ldots\ldots$$

BIENVENIDO T. MIRANDA

UNIVERSITY OF WASHINGTON
SEATTLE, WASHINGTON

[*Editor's Note:* We have usually assumed that a reaction is a tangible process of some sort, rather than a group of words or symbols. If so, one could scarcely

write a *reaction* but rather a *description* of one. If such a description is what we are to mean by the word, "reaction" we can conceivably follow the suggestion made above, but it seems likely to lead to confusion. Only a few of the characteristics of a reaction are described by the usual chemical equation; always the weight balance (if the equation is correct and complete), infrequently the mechanism.]

To the Editor:

It seems to me that Mr. Miranda's questions are really answered by a careful rereading of my paper. However, I offer herewith a set of directions which I hand to students as a guide in applying the method to balancing redox equations.

I. Products of the reaction *must* be known. This information may be obtained from general chemical knowledge but it is fundamentally experimental information.

II. Write all ionic substances as ions with proper charges. Write all weak electrolytes or nonelectrolytes as neutral molecules.

III. Separate the reaction into an oxidation part and a reduction part. Which is which need not be known at this point.

IV. Balance each part separately.

A. *First*, balance atomically, ignoring charges.

1. Balance atoms *other than hydrogen and oxygen* by inspection.

2. Balance hydrogen and oxygen last as follows:

a. in acid solution

(1) if hydrogen is needed, add H^+.

(2) if oxygen is needed, add H_2O and get H^+.

b. in neutral or alkaline solution

(1) if hydrogen is needed, add H_2O and get OH^-.

(2) if oxygen is needed, add OH^- and get H_2O.

B. *Second*, balance electronically by *simply counting net charges on both sides* and adding electrons to the proper side so that there are the same net number of charges on both sides. The half-reactions are now completely balanced. The oxidation half-reaction is the one with electrons on the right side; the reduction half-reaction is the one with electrons on the left.

V. The final balanced ionic equation is obtained by multiplying each half-reaction by an appropriate constant and adding them together such that the free electrons cancel out.

From IVB it is apparent that the decision as to whether a half-reaction is an oxidation or a reduction is a *result to be found*, not a decision arrived at in the beginning arbitrarily.

I do not understand what Mr. Miranda means by "His starting with H in NH_4CNS . . ." It is apparent from the paper that I started with nitrogen. To be sure, exactly where one starts is arbitrary but in general a starting point is indicated in IVA.

I do not propose to try to "explain" Mr. Miranda's equation showing the oxidation of HCl by $KMnO_4$. I propose to arrive at a balanced equation only, if the formulas of the reactants and products are given. If one follows the above directions, writing strong electrolytes as ions, one easily arrives at the half-reactions:

$$2Cl^- = Cl_2 + 2e^-$$
$$MnO_4^- + 8H^+ + 5e^- = Mn^{++} + 4H_2O$$

yielding the balanced ionic equation

$$2MnO_4^- + 10Cl^- + 16H^+ = 2Mn^{++} + 5Cl_2 + 8H_2O$$

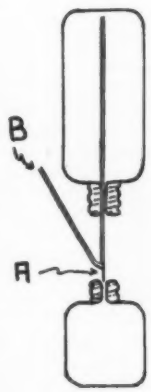
Adding six chloride ions and two potassium ions to both sides and combining ions into neutral "molecules" yields Mr. Miranda's "molecular" equation.

Lastly, it needs to be emphasized that this method is in no way intended to convey meaning as to mechanism. It is only a systematic and essentially infallible technique for arriving at a final balanced equation which in no way depends on arbitrarily assigned oxidation numbers.

D. F. SWINEHART

UNIVERSITY OF OREGON
EUGENE, OREGON

To the Editor:



May I take this opportunity to suggest a solution to the problem presented by Julius Sumner Miller [J. CHEM. EDUC., **29**, 634 (1952)]?

Bend a length of glass tubing (whose diameter is sufficiently small to allow passage of the fluid between it and the bottle neck) at a sharp angle and draw out a small knob (*A*) at the bend as illustrated. With the large bottle upright and with the operator's finger over the opening at *B*, the tube is inserted to the bottom of the large bottle. The whole assembly is then inverted and the flow of air into the large bottle regulated with a finger to produce the desired rate of flow. The oil should flow down the tube and drop off the knob which, of course, is held over the smaller bottle.

LAWRENCE E. CONLON

MOORESTOWN, NEW JERSEY

To the Editor:

Could you please refer me to a good organic chemist who would be ready to correspond with me about some interesting problems in organic chemistry?

D. B. ISHAY

29 HATIKVAH STREET
KIRYAT MOTZKIN
HAIFA, ISRAEL

• PRINCIPLES AND METHODS OF CHEMICAL ANALYSIS

Harold F. Walton, Department of Chemistry, University of Colorado. Prentice-Hall, Inc., New York, 1952. ix + 435 pp. Illustrated. 14.5 × 22 cm. $6.50.

This book is well designed as an advanced text in the more classical forms of quantitative analysis, or, as the author puts it, in "conventional" analysis. Considerable attention is given to the newer methods such as ion exchange, the "complexones," and nonaqueous titrations. Noninstrumental methods, in the present-day usage of the term, are stressed, although the author realizes the difficulties of such a distinction. Thus coulometric methods and high frequency titrations are included, and potentiometry is thoroughly discussed as being germane to all titration procedures. The general plan is to discuss the fundamentals of the various methods and then to present practical examples taken from useful procedures and, in some cases, the techniques involved. Problems are included in all but the introductory chapters. The emphasis is not on the procedures or on explicit directions; however, a wide variety of determinations are mentioned. Enough practical information is contained in the book that its value as a general reference is enhanced. The text presupposes a knowledge of physical chemistry in the development of the theoretical parts, and considerable organic chemistry is included throughout. The book fills a need for texts or references between the usual undergraduate material and more specialized graduate-level courses. It would fit best as a senior undergraduate test, but since our programs do not generally allow room for such a course, it can serve in a beginning graduate course.

Roughly the first half of the book treats gravimetric analysis and separations. This includes electrolytic methods (with coulometric work incorporated here), organic precipitants, complex ions (especially as applied to separations), ion exchange, distillation, and solvent extraction. The remainder is devoted to volumetric analysis with a somewhat unusual arrangement of the topics. After a brief introduction, oxidation-reduction potential theory is introduced, and the methods of potentiometric titrations are discussed. Acid-base titrations, including a brief review of nonaqueous solvents, and acid-base indicators are then presented. Oxidation-reduction methods from the point of view of the specific reagents, applications of oxidation-reduction in organic chemistry (including the Karl Fischer reagent), and oxidation-reduction indicators are treated in order. Precipitation methods, from the point of view of the indicators, and complex-forming titrations, including chelating agents or "complexones," are then discussed. Finally, a chapter on conductometric titrations, including brief descriptions of high frequency and amperometric titrations, is given. The appendixes include a bibliography of texts, a ten-page table of selected methods for determining the elements, numerical data, and answers to some of the problems. The material is conveniently documented, particularly for the period 1940–50, although later as well as earlier references are given.

The number or errors in printing seems high, some of them of a troublesome nature. Thus the potential of the chromous-chromic couple has the wrong sign, a necessary pH on page 319 is missing, a reaction with $LiAlH_4$ is not balanced, a potentiometric titration of $LiAlH_4$ uses two reference electrodes, and acetic anhydride and benzaldehyde are confused in a reaction on page 326. The reviewer objects somewhat to the all-inclusive implication of the statement "...which we shall use as an example to show how all acid-base indicators work" appearing on page 248. The subsequent discussion on the origin of color in indicator systems is in error in attributing absorption to the "oscillation" of electrons among resonance forms. The final chapter is somewhat more difficult to justify in the field of noninstrumental analysis than the other topics, but it is well written and defended on the basis of indicating newer methods as well as other techniques in some difficult systems. The only topic which the reviewer feels was slighted is a more detailed discussion of the colloidal state. Much of this material is implicit in the treatment, but the mechanism of coagulation and peptization is barely mentioned.

JOHN H. WISE

Stanford University
Stanford, California

• PRACTICAL ORGANIC CHEMISTRY

F. G. Mann, Trinity College, Cambridge, and *B. C. Saunders,* Magdalene College, Cambridge. Third edition. Longmans, Green and Co., London and New York, 1952. xv + 466 pp. 76 figs. 14.5 × 22 cm. $3.75.

Because some teachers of organic chemistry may not be acquainted with earlier editions of this laboratory manual, it seems in order to describe briefly the organization of this third edition.

Part I Methods and Manipulations (46 pages), is a brief but comprehensive description of the more common manipulative processes used in the elementary organic laboratory.

Part II, Preparations (187 pages), contains a varied fare of the common syntheses usually found in most elementary manuals. There are approximately 35 preparations of aliphatic compounds and 65 preparations of aromatic compounds. For the most part these are separated in the text, that is, aliphatic from aromatic. The quantities of reagents used are usually as small as is practicable for the hands of elementary students. A few semimicro preparations are included. Examples of reactions illustrating the benzidene rearrangement, Ullmann condensation, benzilic acid rearrangement, Reformatsky reaction, Clemmenson reduction, Fischer indolization, Mannich reaction, and the Diels-Alder reaction have been added in this revision. The experimental directions are exceptionally clear and concise. Their character prompted Sir William Pope to state in the foreword of the first edition, "The student who fails in any of the prescribed experiments should look for the cause of the mishap in his own disregard of the meaning of the printed word."

Part III, Reactions and Identification of Organic Compounds (85 pages), is an elementary approach to qualitative organic analysis in which no attempt is made to include more than a cursory experience in the identification of organic compounds. The authors have felt that students should be thoroughly familiar with the reactions of the more common classes of compounds before attempting more elaborate procedures. The main scheme involves such tests as pyrolysis with and without soda lime, identification of elements, solubility in acidic and basic solutions and in concentrated sulfuric acid, carbonyl reactions, and reactions with ferric chloride. There is no formal classification as to solubility behavior. The treatment seems quite adequate for use in conjunction with the work in the elementary course.

Part IV, Quantitative Analysis (87 pages), is an excellent presentation of the common procedures used in the analysis of organic compounds. There is one section of macroanalyses and another of semimicro analyses. The latter section was written by Dr. P. Sykes of Cambridge. The section of macroanalysis includes the Carius methods for halogen and sulfur, exceptionally

complete descriptions of several methods for the determination of molecular weights, and estimation of hydroxyl groups, amino groups, acetyl groups, formaldehyde, urea, sugars, and simple amino acids. The section on semimicro analysis includes estimation of carbon and hydrogen, nitrogen (Dumas and Kjeldahl) and methoxyl.

Part V, Simple Enzyme Reactions (18 pages), is a novel feature usually not found in laboratory manuals of this type. Although the material appears to be the same as in the first edition, it seems appropriate to mention the presence of this section in the third edition. Biology and premedical students who are bored by the usual preparations and exercises in the laboratory will find interesting reactions in this section.

The text is probably one of the most adaptable laboratory manuals of its type. It is extremely well done with meticulous regard to detail. Every teacher of organic laboratory should examine this book if he has not already done so.

DONALD C. GREGG

UNIVERSITY OF VERMONT
BURLINGTON, VERMONT

● SELECTED VALUES OF CHEMICAL THERMODYNAMIC PROPERTIES

Circular of the National Bureau of Standards 500. By *Frederick D. Rossini, Donald D. Wagman, William H. Evans, Samuel Levine,* and *Irving Jaffe*. United States Government Printing Office, Washington, D. C., 1952. iv + 1268 pp. 21 × 27 cm. $7.25.

THIS book constitutes a very valuable compilation of the thermodynamic data available up to April, 1950. Its price is so relatively low that any serious worker in the field of chemical thermodynamics can well afford to purchase a copy for his personal library; and, of course, all educational institutions and research laboratories will find it very essential for their libraries.

The present work is largely an outgrowth from "Thermochemistry of the Chemical Substances" by F. R. Bichowsky and F. D. Rossini (1936). It is, however, considerably more comprehensive than the earlier compilation and has been prepared by Dr. Rossini and his four collaborators as a definite project at the National Bureau of Standards. It deals with data for compounds of all the elements, except carbon compounds containing more than two carbon atoms.

First, six pages of introduction involve a brief discussion of units and fundamental constants, symbols, standard states, internal consistency, uncertainties, and the order of arrangement of the tables. The notation is essentially that of Lewis and Randall, and all data are expressed in terms of the defined thermochemical calorie.

Next come the various tables. Those of Series I cover 530 pages and contain the available values for: The heat of formation, free energy of formation, entropy, heat capacity, and logarithm of the equilibrium constant of formation, all at 25 °C., and also the heat of formation at 0 °K. The tables of Series II, occupying over 280 pages, then follow with values for the heat, temperature and entropy of transition, fusion and vaporization. Finally, the authors present references to the tables of both series, as well as an assembly of general references, which add greatly to the value of their work.

The book is easy to use, and it is to be hoped that frequent revisions in the future will keep the compilation up to date. The magnitude of the compiling task has become such that only an organization like the National Bureau of Standards will have the time and adequate resources to undertake it. The user of the present tables will frequently find a blank space where he is seeking a desired value. Such blanks should provide incentives for numerous future researches in chemical thermodynamics.

GEORGE S. PARKS

STANFORD UNIVERSITY
STANFORD, CALIFORNIA

● RESPIRATION IN PLANTS

***Walter Stiles,* Emeritus Professor in the University of Birmingham, and *William Leach,* Professor of Botany in the University of Manitoba. Methuen & Co. Ltd., London, and John Wiley & Sons, Inc., New York, 1952. vii + 172 pp. 10 figs. 15 tables. 10.5 × 17 cm. $2.25.**

THE authors of this pocket-sized treatise on such a fundamental biological phenomenon as respiration have aimed at a discussion of principles rather than a cataloguing of detailed observations. The result is an account of the process which is understandable and useful to both the elementary and advanced student.

After an analysis and evaluation of technical procedures, there follows a brief survey of some of the classical and recently more critical work in a chapter devoted to aerobic respiration and another dealing with anerobic conditions. The fourth chapter is concerned with mechanisms of the process. It occupies nearly half of the book and has been largely rewritten for the present edition. Here the student will appreciate the concise descriptions of the several molecular organizations of glucose involved as well as the discussion of phosphorylation mechanisms and enzyme systems associated with anerobic respiration.

A conservative point of view is maintained in considering the identity of aerobic cycles in plants with the more extensively demonstrated Krebs cycle in animal respiration. The chapter closes with a discussion of the intriguing process of oxidative anabolism and the nature of energy transfer through energy-rich phosphate bonds.

Those of us who are engaged in teaching courses at the general level constantly feel the need for providing students with the opportunity for independent pursuit of some particular phase of the subject. With regard to physiological studies, the elementary student often feels his lack of knowledge of even the rudiments of biochemistry. A book such as "Respiration in Plants" will give the student at this point a very necessary lift in his intellectual endeavors.

GEORGE L. CHURCH

BROWN UNIVERSITY
PROVIDENCE, RHODE ISLAND

● GENERAL AND INORGANIC CHEMISTRY

***P. J. Durrant,* Lecturer in Chemistry, Selwyn College, Cambridge. Second edition. Longmans, Green and Co., New York, 1952. x + 761 pp. 140 figs. 63 tables. 14.5 × 22 cm. $4.50.**

THIS text is, as was true of the first edition published in 1939, "designed particularly for students reading chemistry for higher School Certificate Examinations, for Preliminary Medical Examinations, and for Entrance Scholarships to the Universities." Ten chapters that deal with general principles are followed by 16 chapters devoted to the study of the elements and their important compounds. Forty pages of examination questions that have been selected from universities of the British Commonwealth are included in the book. Answers are given for all of the mathematical problems in these questions. The book has a good index. The majority of the figures are line drawings to illustrate industrial processes for the preparation of chemicals. The tables give an abundance of physicochemical data. Photographs are not included. No exercises or reading lists are given at the ends of chapters. The paper is of good quality, the type is clear, and the book is attractively bound.

This reviewer read the book with both interest and profit. The definitions, statement of laws, and explanations of theory are clearly worded and logically developed. In some instances it is felt that conciseness was achieved at the expense of needed qualifications. Historical material is rarely given. Descriptive chemistry is not included except in a limited way in connection with industrial processes for the manufacture of chemicals.

The first four chapters cover atomic and molecular theories, the states of matter, and molecular, atomic, and equivalent weights.

Numerous ways are shown by which these "weights" have been determined. Crystal structure and isomorphism are thoroughly discussed. It is felt that most American students of first-year chemistry would find the mathematics of these early chapters rather rigorous. Atomic structure is first introduced in Chapter V. The periodic classification of the elements, electronegativity, and valence are all intimately and effectively tied to atomic structure. The periodic chart used in this text has 11 groups and is based on the chart proposed by T. S. Wheeler ["Chemistry and Industry," 1947, pp. 639—42]. Teachers who want a refreshing approach to this area of the first-year course should read this text.

The subject of solutions is treated from the viewpoint of physical chemistry; numerous phase equilibrium diagrams are given. Osmosis and fractional and steam distillations are covered in some detail. The chapter on electrolytes is not up to the high standard of the rest of the book; the definitions are poor and many very important aspects of the subject are not mentioned. No basis is laid for volumetric analysis or equivalency of chemicals. The term "normal solution" is mentioned but not defined. The chapter on electrode potential and electrolysis is excellently developed. The subject of colloids is well handled as regards definitions and classifications but the properties of colloidal solutions are not adequately compared with the solutions of molecular and ionic materials. Modern methods of using ion-exchange resins are not mentioned in connection with water treatment (although zeolite is mentioned).

The chapters that are devoted to the elements and their important compounds all follow the general pattern of giving (1) tables of physiochemical data, (2) generalized statements of the chemical properties as developed from their structure, electronegativity, and periodic classification, (3) industrial processes for the preparation of the elements and their major compounds, and (4) specific chemical properties. Most of the equations are written molecularly. In reading these chapters, it is necessary to keep constantly in mind the periodic chart that is used in this text. General principles are constantly stressed.

It is the opinion of this reviewer that American teachers of first-year college chemistry courses would profit by reading this text. At the same time, he is doubtful that it is a suitable text for most courses in this country.

JOHN B. ENTRIKIN

CENTENARY COLLEGE
SHREVEPORT, LOUISIANA

● INDUSTRIAL AND MANUFACTURING CHEMISTRY. PART I: ORGANIC

Geoffrey Martin. **Revised by *Edward I. Cooke.* Seventh edition. The Technical Press, Ltd., England; Anglobooks, New York, 1952. xxi + 752 pp. 240 figs. 16.5 × 25 cm. $21.**

ARE you interested in the manufacture of candles? Chapter IXA is devoted to this subject. The material given is up to date; 1910, that is.

The reviewer compared this new edition with the fifth (1920) edition and found but few changes. There is still the same chapter on Artificial Silk. The Aliphatic Chemicals Industry is the manufacture of oxalic acid from sawdust, tartaric acid from wine lees, plus a few halogen derivatives and esters. These chapters indicate the age, tone, and level of the book.

Certain revisions and additions have been made, such as a 35-page chapter on the Petroleum Refining Industry, which was fitted between pages 4 and 19 by having page 18 run from 18*a* to 18*l*. Mostly the pages are printed from the same plates used for the fifth and sixth editions. Even without this economy, a price of $21 would be fantastic.

This book outlived its usefulness after the sixth edition in 1922.

KENNETH A. KOBE

UNIVERSITY OF TEXAS
AUSTIN, TEXAS

● MR. WIZARD'S SCIENCE SECRETS

Don Herbert. **Popular Mechanics Press, Chicago, 1952. 264 pages. Illustrated. 14.5 × 22 cm. $3.**

AN ATTRACTIVE selection of simple "home experiments" in general science which should provide profitable entertainment for youngsters on many a rainy afternoon. Chemistry is represented to a reasonable extent. The chapter headings are inviting: Right in Front of Your Nose (air pressure); Holes in the Air (vacuum); What's Burning? (oxidation); "What Makes an Airplane Fly? (aerodynamics); Why Balance Gets Lost (gravity) Getting Warm (heat radiation, conduction, and convection); "Sound Fun (sound); The Bottle, Pin and Soda-straw Philharmonic (musical vibration); The Case of the Mysterious Magnet (magnetism); Electromagnetic Magic (electromagnetism); Charge it! (static electricity); The Wonders of Water (solids, liquids, gas); Bubbles at Work (carbon dioxide); The Biggest Mystery of All—You! (the human body).

● THE SCIENCE OF FLAMES AND FURNACES

M. W. Thring, **Head of Physics Department, British Iron and Steel Research Association. John Wiley & Sons, Inc., New York, 1952. xiv + 416 pp. Illustrated. 14.5 × 22.5 cm. $6.50.**

THIS book is clearly a "must" for engineers of all classifications who are concerned with the design and operation of industrial furnaces, for physicists and chemists interested in theoretical and practical problems of furnaces, and teachers and students of engineering who desire a modern authoritative introduction to the scientific aspects of industrial furnace operations. The author is eminently qualified for his subject, having specialized in furnace research for many years. The subject matter itself is manifestly of vast economic significance and in addition offers interesting problems both in applied and fundamental research: this is a combination of circumstances which might be expected to attract the attention of industrial and other research organizations in ever-increasing measure.

The opening chapter presents thumbnail sketches of the function of furnaces in the production of iron, steel, nonferrous metals, ceramics, chemicals, cement and glass, and in coal carbonization. It proceeds to classify the branches of science applicable to the processes taking place in the furnace, namely (1) the thermodynamics of heating processes, (2) the laws of energy release by combustion of fuels, (3) the laws of flow of hot gases, and (4) the science of construction of furnace walls. These subjects are taken up separately in the succeeding chapters.

In the second chapter entitled The Thermodynamics of Furnace Heating, the knotty problem of standards of efficiency is treated. From the first and second law the applicable concepts are developed step by step to the ultimate efficiency standard based on consideration of the "virtue" of energy, a quantity derived from the second law which is conveniently used here in place of entropy.

In logical order, the next chapters deal with the various aspects of conversion and transfer of energy in the furnace. Chapter 3 concerns the liberation of heat by combustion; this includes reaction energies and equilibria, reaction kinetics, flames of premixed gas and diffusion flames, combustion of solid fuels and liquid fuel sprays, and the means of control of combustion. Chapter 4 deals with heat transfer by conduction, convection, and radiation; the means of measurement and calculation of heat transfer in the furnace chamber, and of heat loss through the furnace "skin"; the unsteady state during the heating-up period. Chapter 5, entitled The Aerodynamics of Hot Systems is of particular interest as it deals with a comparatively little-known subject. Its significance in furnace operation arises from considerations of the quantity of flowing gas, and the velocity distribution and stratification of hot and cold gas in the stream. The text discusses the aerodynamic principles involved, methods of observation and measurement, and results obtained.

Following this coverage of the dynamic aspects of furnace

operation, Chapter 6 reviews the science of furnace construction—the testing and properties of refractories, the construction principles for wall and crown. The last chapter summarizes the application of the scientific method to furnaces. This is given under three main topics: diagnosis of a given single furnace, statistical study of a furnace type, and modification of an existing type. Acknowledgment is made to the fact that the complexities of furnace design and operation make it necessary for science to work in partnership with "art" or "know-how."

A table of conversion factors and an index conclude the book; most chapters are followed by individual summaries.

It is to be hoped that this book will become widely distributed throughout industry, research institutions, and engineering schools.

GUENTHER VON ELBE

U. S. Bureau of Mines
Pittsburgh, Pennsylvania

• SOLUBLE SILICATES: THEIR PROPERTIES AND USES (2 VOLUMES)

The late *James G. Vail,* Director, and *John H. Wills,* Research Supervisor of Patents, Philadelphia Quartz Co. A. C. S. Monograph No. 116. Reinhold Publishing Corp., New York, 1952. Volume 1 ("Chemistry"): xii + 357 pp. Illustrated. 15.5 × 23.5 cm. $9. Volume 2 ("Technology"): xxi + 668 pp. Illustrated. 15.5 × 23.5 cm. $15.

This work, which replaces Dr. Vail's long-out-of-print A. C. S. Monograph No. 46 on "Soluble Silicates in Industry," is described on the jacket as "the only complete treatment in any language." No one will argue with this appraisal, biased as it may be, because the amount of material covered by its 931 pages of text, its 2300 references, and its 90 pages of index can only be described as staggering. If you would like to develop photographs, build rubber roads in Malaya, study the phase relationships in the quaternary system Na_2O—CaO—SiO_2—H_2O, grow large crystals of quartz, or do anything else in which soluble silicates will be useful, this is your book.

Volume 1 makes a great effort to collect all the theoretical and physicochemical information on soluble silicates in one place, classified in five chapters concerning history, present practices in preparation, homogeneous systems, heterogeneous systems, and complex systems. The impressive amount of information obtained at the Geophysical Laboratory of the Carnegie Foundation is incorporated through the active cooperation of Dr. George W. Morey, and similar sources well known to Dr. Vail have been enlisted to great advantage. This factual information on the silicate systems would be an indigestible mass of data were it not for the almost complete translation into graphical form. The authors and the staff of Philadelphia Quartz Co. have gone to a great deal of trouble to present a critical survey in a readable form, and the numerous figures and tables are a great help.

Just as personal impressions of Volume 1, your reviewer noted with particular interest the discussions of the conservation of proteins, starch, and scarce phosphates by the substitution of sodium silicates, the analytical procedures given in full, the complete treatment given to surface tension and viscosity of solutions, the crystallographic and petrographic data, and the striking photomicrographs. Despite the separation of chemistry from technology in this volume, the utilitarian aspects are always pointed out (as in the discussion of water softening in Chapter 5). This is a very practical book.

Volume 2 presents, in the words of Dr. Wills, "some theoretical statements designed as a guide to practical use of soluble silicates in particular applications but, in the main, describes where and how the soluble silicates have been found valuable in the past, especially in the more recent years since the publication of the original monograph." The information is divided into chapters on interfaces modified by silicate solutions (detergents and detergent mixtures, effects on metal and other inorganic surfaces, effects on textile fibers and other organic surfaces, dispersion and complex applications), coatings and films (protective and decorative coatings, preservation of stone and other porous materials, sizing and impregnation), bonded surfaces (adhesive films, conjunctive cements, consolidated masses, stabilization of soils), sols, gels, and polymers (fluid sols, coagulation, dry gels, catalysts), and physiological behavior (internal effects, external effects, pharmaceuticals). The thousands of entries in the author index and the 45 pages of subject index make it easy to look up a certain topic, but your reviewer was constantly distracted by the neighboring accounts of such things as the use of silicates on the aforementioned Malayan roads, in paper boxes in which gasoline may be burned, in the arc cutting of metal under water, in drawing bolts, in making television tubes, in de-inking paper, and in washing tripe. He is convinced now that he never knew anything about sodium silicate, but is glad that the illustrations and tables of this volume are there to help him learn at this late date.

Dr. Vail has long been appreciated by chemists the world over, but even if he had not been, his name would henceforth be blessed by the thousands of chemists who find they need to know something about the theory or practice of soluble silicates and are saved many hours of digging in the original literature by this really expert work.

E. G. ROCHOW

Harvard University
Cambridge, Massachusetts

• ULTRAVIOLET RADIATION

Lewis R. Koller, Research Associate, General Electric Research Laboratory. John Wiley & Sons, Inc., New York, 1952. ix + 270 pp. Illustrated. 15 × 23 cm. $6.50.

This book is a good source of technical information concerning the production, measurement, and selected applications of ultraviolet light. A detailed description of arcs and other laboratory sources is given, with many illustrations, spectral distribution curves, and tables of operational characteristics. The remainder of the book is concerned mainly with discussion of selected topics, which are: solar radiation and its ultraviolet content, the transmission and reflection of materials, methods of detection of ultraviolet radiation.

The principal applications are to ultraviolet therapy and germicidal effects, the production of ozone, and to fluorescent radiation. No attempt is made to treat all applications comprehensively.

The technical descriptions are clear and well written. This reviewer found the information on arc characteristics most valuable, and recommends the book to others who use sources of ultraviolet radiation.

A. B. F. DUNCAN

University of Rochester
Rochester, New York

• INK AND PAPER IN THE PRINTING PROCESS

Andries Voet, Director of Ink Research, J. M. Huber Corporation, Borger, Texas. Interscience Publishers, Inc., New York, 1952. xii + 213 pp. 80 figs. 15.5 × 23.5 cm. $5.90.

The author of this book came as a guest to this country in 1935; he returned in 1939 to stay, attain citizenship, and become a leading scientist in the printing ink industry. Called upon to lecture on ink technology at New York University, he found that recent reports of scientific investigations in the field (including a number conducted in his own laboratory) were not readily accessible to students. The book is offered, therefore, as an introduction to the physics and chemistry of ink and its interaction with paper. It is intended for somewhat advanced students of the graphic arts who should have a basic understanding of scientific principles, and who are presumed to be familiar with general formulation and technology, color, gloss, and dispersion.

These aspects are therefore omitted, in favor of a basic review of the rheology of inks and the principles governing their behavior on the printing press, their transfer to paper, and their subsequent drying on the printed surface. On the whole, the book fulfills this mission well, and is a useful contribution to ink literature. The author is to be congratulated on proving his point that the art of ink making is gradually maturing into a science, though still far from this final goal.

Part I of the book, 86 pages, deals with the behavior of ink on the press up to the instant of impression. The all-important flow properties of inks and the modes of separation of films at the various points of contact between rollers, printing surface, and paper are fully discussed, with mathematical treatment and numerous graphs wherever warranted by known data. Instrumentation and methods of measurement are described. The author's claim that viscoelastic behavior explains high-speed film splitting seems well supported, but will be challenged in some quarters. Causes of, and some ways of relieving, troublesome "ink flying" are considered.

Part II, 118 pages, goes into the relationships of ink and paper: halftone reproduction, contrast and coverage, penetration, print-through, ink transfer, picking, offset, and drying by the various physical and chemical processes. As is natural, the viewpoint here is that of the ink, rather than of the paper technician; one could wish that something more of the possibilities of adaptation of paper to ink, rather than the reverse, had been brought out.

The book is very well written in the main, but there are a few obscure or incorrect statements. The one on page 60 that in multicolor printing each impressed film must have a more pronounced resistance to film separation than the one immediately preceding it, is properly contradicted on page 147. And on page 192, Dr. Voet hardly meant to say that "prints of 3 micron thickness did not require a longer drying time than inks of about twice this film thickness." For his oxygen availability test at this point, he would undoubtedly use his glass plates to *confine* prints between unprinted sheets, rather than to "separate" them. There are very few typographical errors.

CHARLES R. BRAGDON

LARCHMONT, NEW YORK

● TEXTILE CHEMICALS AND AUXILIARIES

Edited by *Henry C. Speel,* Consulting Chemist, R. S. Aries and Associates, New York, N. Y. Reinhold Publishing Corp., New York, 1952. v + 493 pp. 45 figs. 35 tables. 16 × 23.5 cm. $10.

ANYONE interested in gaining an introductory knowledge of the textile industry, the chemical industry's best customer, will find this book an excellent place to start. The book is divided into two parts, and the 142 pages of Part I constitute the best short introduction to textile science to come to the attention of the reviewer. It is recommended to the teacher of home economics as well as the general chemistry teacher, for it covers the whole field of textile science, not just chemistry. The information is up to date and includes data on many of the new synthetic fibers, foreign and domestic. Some of the theories are presented as if they were universally accepted, although textile chemists are actually still undecided between alternate theories.

The 15 chapters of Part II discuss the "Raw materials in fabric processing," covering the well-known materials such as water and starch, as well as the more specialized substance like cationic finishing agents and mothproofing.

The coverage in each field is adequate, written for the chemist, not the specialist in the particular field. The fact that trade names are so frequently mentioned will be of value to many workers. It should be noted that the names given are always those used in industry, not those known to the retail trade, so the teacher of home economics will be helped less than the worker in the textile industry.

As in any book written by various authors, differences in arrangement and emphasis exist between the chapters, but the editor has done an excellent job of keeping them at a minimum. The bibliographies at the ends of the chapters are well chosen and should be entirely adequate for those who wish to go more extensively into any subject.

WILLIAM G. CHACE

LOWELL TEXTILE INSTITUTE
LOWELL, MASSACHUSETTS

● A LABORATORY MANUAL OF PHYSIOLOGICAL CHEMISTRY

D. Wright Wilson, Benjamin Rush Professor of Physiological Chemistry, University of Pennsylvania. Seventh edition. The Williams & Wilkins Co., Baltimore, 1952. 293 pp. 15.5 × 23.5 cm. $3.25.

IN THIS new edition of Wilson's laboratory manual (the first edition was published in 1928) a number of experiments previously included have been omitted, some experiments have been modified, and several new experiments added. The section on the cell nucleus, contributed by Walter Jones, has been retained. Quantitative photometric experiments are described for the Klett-Summerson instrument. The author continues to state that the manual "is intended to be used as a teaching manual and not as a comprehensive reference book.... we have found no difficulty in using this manual in our medical, dental and veterinary courses."

The book is still printed on one side of the paper, the right-hand page remaining blank for note-taking. This practice has borne much criticism in the past. It is this reviewer's feeling that where a series of short experiments involve only a brief procedure and one or possibly two results are concerned a blank page bound in the text offers many advantages over the notebook write-up. In such experiments the student finds it convenient for quick reference and study to paraphrase the experiment into one or two brief sentences; in some cases schemata are valuable; and often a chemical equation may suffice. The blank page opposite the text of the experiment is ideal for this purpose.

The first 40 pages (actually 23 pages of text) are devoted to a review of qualitative tests for certain inorganic constituents, reactions of certain organic groups, standardization of acid and base, electrolytic dissociation theory, and colloids. Some of this material could be included with profit in the sections where it applies, while the quantitative analysis theory and practice might well be left in texts covering that material. The student has presumably been led through a course in quantitative analysis and either owns texts on the subject or is in a position to consult them effectively. There could be a tendency on the part of the student to wonder why he must perform 23 pages of preliminary work. He is in all probability discussing carbohydrates, fats, and proteins in lecture during this period. Assuredly, this section might be omitted entirely or in part, or assigned as review without performance.

The sections on carbohydrates, proteins, and fats include the standard procedures. Composition and directions for preparation of the required reagents are included as footnotes. Occasional questions are asked of the student and it would seem that more of this would prove beneficial. A quantitative determination of liver glycogen is given. The section on proteins includes preparation of a crystalline globulin and cystine and tyrosine by acid hydrolysis of hair (the excess acid is neutralized) but there is no preparation of an amino acid by enzymatic hydrolysis. No mention is made of the ninhydrin reaction at this point though the reaction is used later. The iodine number is determined by the Rosemund method which employs bromine in the form of pyridine sulfate dibromide. The section on lipids also includes an experiment on paper chromatography of a sample of crude phosphatide hydrolysate. The experiment is lengthy (12–14 hours for the separation of the components on the paper) and offers no great advantage over the simpler amino acid separation which can be adequately performed in a laboratory period.

The second part of the book (pages 92–290) includes experiments on digestive processes, milk, qualitative and quantitative procedures on blood and urine. Following this, a page is devoted to the statement, "the use of tagged compounds will be demonstrated and the student will have the opportunity of observing the use of the Warburg apparatus and the use of the Geiger-Müller counter assembly for the incorporation of radioactive glycine into hypoxanthine by the enzymes present in the liver of pigeons."

The book is well bound in hard covers, opens well for use in the laboratory, and the typography is excellent.

LIONEL JOSEPH

San Diego State College
San Diego, California

• THE CHEMISTRY OF SYNTHETIC DYES. VOLUME II

K. Venkataraman, **Director, Department of Chemical Technology, University of Bombay. Academic Press, Inc., New York, 1952. xv + 738 (705–1442) pp. Illustrated. 15 × 23 cm. $15.**

This volume contains the final 20 chapters of the work, the first volume of which was reviewed recently [J. Chem. Educ., 29, 426 (1952)]. Author, subject, and dyestuff name indexes for the entire treatise and a Table of Errata for Volume I are included. Pagination of both volumes is continuous.

The high quality of the first volume is maintained. Professor Vekataraman concludes the survey of dyes following the chemical classification adopted in Volume I. The discussion of sulfur dyes alone requires 58 pages. This is followed by chapters on the phthalocyanines, the cyanine dyes, the action of light on dyes and dyed fibers, the relationship between chemical constitution and substantivity, and the identification, analysis, and evaluation of dyestuffs.

As stated before, this treatise is without doubt the best work on dyes to date, covering the field thoroughly to about 1950.

EDWARD R. ATKINSON

Dewey and Almy Chemical Company
Cambridge, Massachusetts

• THIOPHENE AND ITS DERIVATIVES

Howard D. Hartough, **Socony-Vacuum Laboratories, Paulsboro, New Jersey, with special chapters by *F. P. Hochgesang* and *F. F. Blicke*. Interscience Publishers, Inc., New York, 1952. xvii + 533 pp. 27 figs. 107 tables. 15.5 × 23.5 cm. Single volume, $16.50; subscription price, $15.**

The chemistry of heterocyclic compounds is important and complex, but still not well organized. In this volume the author has performed a real service to those interested in heterocyclic compounds by collecting and organizing most of the published information concerning thiophene and its derivatives. This tremendous task has resulted in a book which is accurate, complete, and readable.

The importance of thiophene has grown steadily since its discovery by Victor Meyer in 1882. Two previous summaries of thiophene chemistry in 1888 and in 1941 (both in German) attest to a steady extension of our knowledge of thiophene and its compounds. In recent years this growth has been exponential, resulting in much new material being brought together for the first time in this book.

The purpose of the author was to include every significant published reference to thiophene and its derivatives. In addition there is a complete summary of the unpublished data on thiophene obtained in the Socony-Vacuum Laboratories. Toxicological and pharmacological properties of thiophene are reviewed by Dr. F. F. Blicke. Spectrochemical and related properties of thiophene are covered by F. P. Hochesag. More than half of the book deals with the synthesis, physical properties, and reactions of thiophene, its homologues, and its functional and nonfunctional derivatives. Whole chapters are devoted to halothiophenes, nitro- and aminothiophenes, hydroxythiophenes, aldo and keto derivatives, carboxy derivatives, sulfur derivatives, etc., of thiophene. Other chapters deal with substitution reactions in thiophene nucleus, selenophene and tellurophene, and laboratory preparations of thiophene compounds.

The information in the book is presented in a thorough, exact, and attractive manner. Experimental results are critically evaluated and organized about basic principles. The use of many tables, graphs, and structural formulas contributes to the readability of the volume. High-grade paper and typography give it a pleasing appearance.

This volume will be very helpful to anyone interested in heterocyclic chemistry, and in particular thiophene chemistry. It is a convenient source of information concerning the synthesis and properties of hundreds of specific derivatives of thiophene. Actual laboratory directions for the preparation of important intermediates will be helpful to those engaged in experimental work in the field. Students and teachers will find here a thorough treatment of the principles of thiophene chemistry. Research chemists and engineers will find it a useful reference for the preparation and identification of thiophene derivatives. Pharmaceutical chemists will find it helpful because of the importance of the physiological activity of the thiophene nucleus in modern drugs.

ERNEST M. HODNETT

Argonne National Laboratory
Lemont, Illinois

• VOLUMETRIC AND PHASE BEHAVIOR OF OIL FIELD HYDROCARBON SYSTEMS

M. B. Standing, **Division Supervisor of Drilling and Production Research, California Research Corporation. Reinhold Publishing Corp., New York, 1952. vi + 123 pp. 69 figs. 35 tables. 22 × 28.5 cm. $10.**

Few books have been written concerning the behavior of hydrocarbon systems in the natural state in underground oil and gas reservoirs. This volume will be welcomed by professors and practicing engineers alike as a valuable aid in determining fluid properties and predicting the behavior of oil-field hydrocarbon systems. The routine analyses made in the petroleum laboratory on samples of reservoir fluids are discussed, together with the application of laboratory data to the solution of reservoir engineering problems. The results of the author's own research have been included along with material from many technical articles.

This book is written mainly for the petroleum engineer, geochemist, and geologist, although certain sections could very well be adapted to classroom use. As a textbook it has two definite advantages: (*a*) the exposition of technical details is very lucid; and (*b*) the terminology employed by the author is consistent with that in general use by petroleum and reservoir engineers throughout the oil industry.

Discussions of phase behavior have been divided into three general categories: gases, condensate systems, and dissolved gas systems. Most readers will probably find the sections on dissolved gas systems of paramount interest because of the greater frequency of this type of petroleum accumulation. In Chapters 7 and 8 the laboratory tests on dissolved gas systems are described in detail, and the proper utilization of laboratory data in volumetric-balance equations is indicated. Due recognition is given to the increasing economic importance of condensate reservoirs by including a chapter on the behavior of gas-condensate systems (Chapter 6). Other chapters are entitled: Introductory Concepts, Sampling Methods and Apparatus, Behavior of Gases, Behavior of Liquids, and Vapor-liquid Equilibria. Included as an appendix are three charts (separately printed) to be used in estimating reservoir fluid characteristics from the properties of surface production.

This book is a valuable contribution to the literature of petroleum reservoir engineering. A better understanding of the behavior of hydrocarbon systems will permit more accurate evaluation of oil and gas reserves and will aid in the constant efforts of petroleum technologists to improve production practices.

A. G. WEBER

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● PLANTS OF THE BIBLE

Harold N. Moldenke, Curator and Administrator of the Herbarium, New York Botanical Garden, and _Alma L. Moldenke,_ Biology Department, Evander Childs High School. Chronica Botanica Co., Waltham, Mass., 1952. xix + 364 pp. 95 figs. 17.5 × 27 cm. $7.50.

IN A BOOK so universally known as the Bible, it is highly desirable that accuracy in reference to characteristics of the ancient Palestinian scene be extended to correct identification of the plants and animals involved. Interest in the plants of the Bible is evident from the fact that many publications on the subject have appeared in recent years. Probably none has been prepared with more scholarly effort and greater authority than "Plants of the Bible," a work which is the product of twelve years of preparation by Harold N. Moldenke and his wife.

In this period, nine revised or new versions of the Bible by eminent scholars have appeared—all these in addition to the several versions in the English language that have been printed since the King James version in 1611! When it is realized how greatly various versions may differ in the interpretation of plant references alone, the magnitude of the task that confronted the present authors becomes apparent. A striking example of confusion in identity of a plant through the vicissitudes of translation is cited by the author in the case of Nahum 2:3. "This is rendered by the King James version—'and the fir trees shall be terribly shaken,' by the Jastrow version—'and the cypress spears are made to quiver," by the Goodspeed version—'and the charges will prance,' by Moffat—'and their horses prance at the muster,' and by the Douay version—'and the drivers are stupified.'" It is agreed that the "fir" mentioned in the Bible was really the Aleppo pine. In the above passage, however, the reference is actually to horses or their drivers.

Among the many causes which have contributed to confusion in interpreting biblical texts is the well-known fact that many of the Old Testament passages were written by men of limited vocabulary from information handed down by word of mouth for centuries. Then too, it was obviously an act of heresy for scholars during the Middle ages to suggest changes in supposed canonical texts. Furthermore, no scientific study of the flora of the Holy Land was attempted until the middle of the eighteenth century when two students of the famous Linnaeus sent him some specimens collected during travels that proved to be quite hazardous. And finally, as the authors of the text indicate, many exotic plants have crept into the region already depleted of some of its native flora by the ravages of careless agricultural practices and subsequent errosion. At the present time, botanists in Near East universities are engaged in a systematic study of this region which has been neglected from the standpoint of its natural history.

Dr. Moldenke, a distinguished plant taxonomist, has appraised the findings both of scientists and men of letters concerned with studies of biblical texts. He and his wife have produced a most valuable book which lists some 230 plants, each with all of the biblical passages concerned. Included are extensive discussions on the etymology of the Hebrew or Greek words in the original texts. The illustrations are both in half-tone and line drawings. There are over 600 bibliographical references. Certainly this volume will be a useful addition to any collection of reference works.

GEORGE L. CHURCH

BROWN UNIVERSITY
PROVIDENCE, RHODE ISLAND

ERRATUM

IN THE review of Martell and Calvin's "Chemistry of the Metal Chelate Compounds" (Preutice-Hall, Inc.) on page 215 of the April issue, the trade price of $13.50 is listed. The price given should have been the college list price, which is $10.

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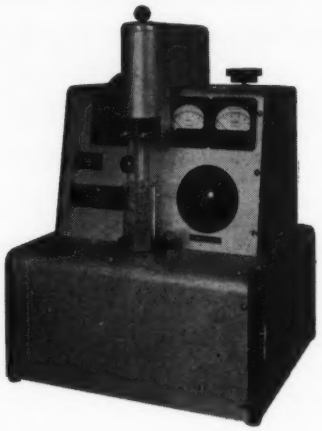

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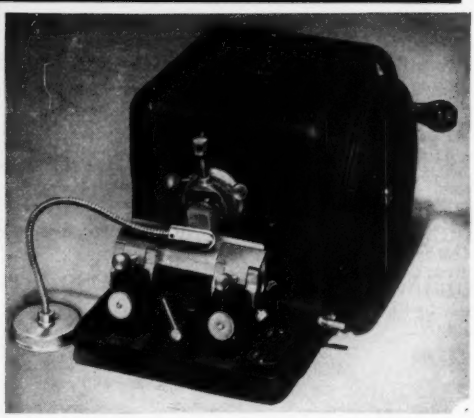

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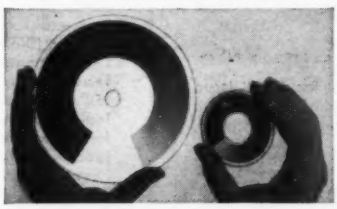

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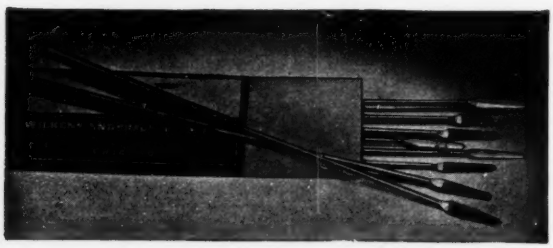

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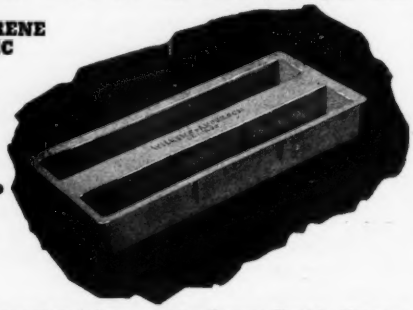

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Out of the Editor's Basket

New Oil Diffusion Pump

One of the most modern developments in oil diffusion pumps is the new Cenco Supervac OD-25. This unit features a radical departure from conventional design with the result that it is exceptionally easy to take apart for cleaning and may be quickly connected or disconnected.

The unit consists of only three basic parts: the jet manifold, the jacket, and the boiler with its detachable heater. The jet manifold, which is a three-stage unit of welded steel, fits snugly in the boiler compartment and is completely enclosed by the jacket. Six Allen screws hold the jacket and boiler together and a synthetic rubber "O" ring provides a vacuum-tight seal.

The Cenco Supervac OD-25 Oil Diffusion Pump is based on the principle of entrapping gas or air molecules by a stream of oil vapor, the oil being vaporized by the application of heat. As oil molecules leave the surface of the oil in the boiler, they are forced upward through a vertical column where they are deflected downward and outward at sonic velocities through jets placed at three different levels. The air or gas molecules from the system are swept along by the oil vapor stream to the lower chamber where they accumulate and are readily removed by the forepump. The oil vapor condenses on the cool surface of the jacket and returns to the boiler.

Full details about this new pump may be obtained by writing the manufacturer, Central Scientific Co., 1700 Irving Park Rd., Chicago 13, Illinois.

New Chemicals

● In addition to the recently announced O.S. (Optically Standardized) Amino Acids, Schwarz Laboratories, Inc., 230 Washington St., Mount Vernon, New York, now has available the following optical isomers of the same grade: l-Cysteine·HCl·H_2O c. p.; l-Leucine O.S. (Methionine Free); l-Phenylalanine O.S.; d-Phenylalanine O.S.; l-Serine O.S.; l-Tyrosine O.S.

● Following are ten new chemicals just added to the list of the Eastman Kodak Co.: Acrylamide; 4-Amino-2,6-dimethylpyrimidine; 2,5-Dimethylfuran (Pract.); 5,5-Dimethyl Hydantoin (Pract.); 2,5-Diphenyloxazole; Formamide; Malonic Acid (Pract.); Phthaloyl Chloride (Pract.); Y-Valerolactone (Pract.); *m*-Xylene (Pract.).

The chemicals are available from the Eastman Organic Chemicals Department, Distillation Products Industries, Rochester 3, New York.

● Bios Laboratories, Inc., 17 W. 60th St., New York 23, New York, announces the availability of the following new chemicals: Acetaldoxime (Acetaldehyde Oxime); 3-Acetylpyridine; Adonidin; Agmatin Sulfate (1-Amino-4-guanidobutane Sulfate); l(+)Carnosine (β-Alanylhistidine); Chondroitinsulfuric Acid; *dl*-Glyceraldehyde; Retene (1-Methyl-7-isopropylphenanthrene); Veratric Acid (3,4-Dimethoxybenzoic Acid); Xanthosine (Xanthine riboside).

● Carbobenzoxy Chloride (Benzyl Chloroformate) is now produced on a commercial scale by Mann Reserach Laboratories, Inc., 136 Liberty St., New York 6, New York, in a minimum purity of 99%.

This reagent is the standard starting material for the synthesis of peptide and peptide intermediates and is also used for blocking of amino or hydroxyl functions generally.

New Literature

● A general catalogue of their Chemical Instruments Division has just been published by Consolidated Engineering Corp., 300 N. Sierra Madre Villa, Pasadena 15, California. Besides illustrating and describing various mass spectrometers and accessories, the catalogue contains a short article on the mass spectrometer principle.

● The Metalab Equipment Corp. announces that their new catalogue and manual No. 4B is ready for mailing to companies, hospitals, schools, and research organizations. It is a complete volume of 180 pages illustrating an extensive line of

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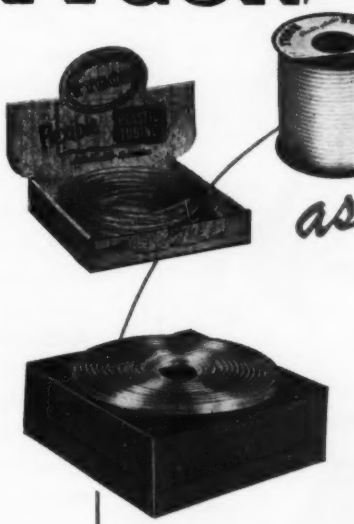

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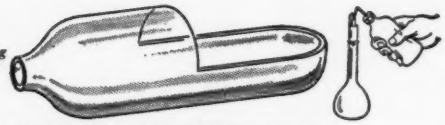

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In addition to listing the Metalab line, the catalogue contains technical information and data of value to directors and technicians planning new laboratories and modernizing old ones. Prominent mention is made of the free engineering and design services available to users of Metalab equipment, including a complete analysis of space economy; counsel on utilization of new equipment and present facilities; ease and coordination of laboratory operation; location and types of plumbing and electrical utilities; ventilation; lighting; and functional design.

Copies of the new catalogue may be obtained by writing on company letterhead directly to the Metalab Equipment Corp., Hicksville, Long Island, New York.

● An inclusive 28-page catalogue of Beckman ultraviolet and visible spectrophotometers—picturing all sample cells and other accessories—has just been released by Beckman Instruments, Inc.

The two-color brochure contains detailed descriptions and illustrations of the Model B and DU Spectrophotometers—precision instruments which measure and identify substances by passing light through them. Such well-known auxiliary units as the Flame, Reflectance and Fluorescence attachments also are described. These multiply the analytical uses of spectrophotometry without requiring a heavy outlay for new basic instruments.

The catalogue features an extensive section treating the complete line of Beckman spectrophotometer cells, interchangeable sample compartments and cell adapters. A composite price list-index is also furnished. Copies can be obtained from Beckman Instruments, Inc., South Pasadena 1, California, through requests for Bulletin 303-36.

● A new 28-page brochure, "Celite—The Story of Diatomite," has just been published by Johns-Manville. Designed for both laymen and technical men, it uses photographs and drawings as well as text to trace the development of diatomite.

The story begins with the arrival of the diatom during the Miocene Era. This first section tells what a diatom is and explains how these marine organisms emerged as the unique deposit in California known as the White Hills of Lompoc. The next section deals with discovery of this deposit and the experiments which have led to the still-multiplying uses of diatomite today.

The succeeding sections of the brochure describe the current benefits of diatomite—as filter powder for water purification, dry cleaning, and the production of foods and beverages; as a mineral filler in the manufacture of paint, plastics, paper, fertilizers, and insecticides; as a high-temperature insulation; and, as a catalyst carrier wherever silica is indicated. The story concludes with a brief prediction of new things that may be expected. Copies of "Celite—The Story of Diatomite" are now available from Johns-Manville, 22 E. 40th St., New York 16, New York.

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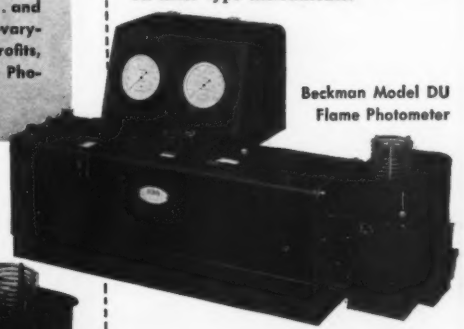

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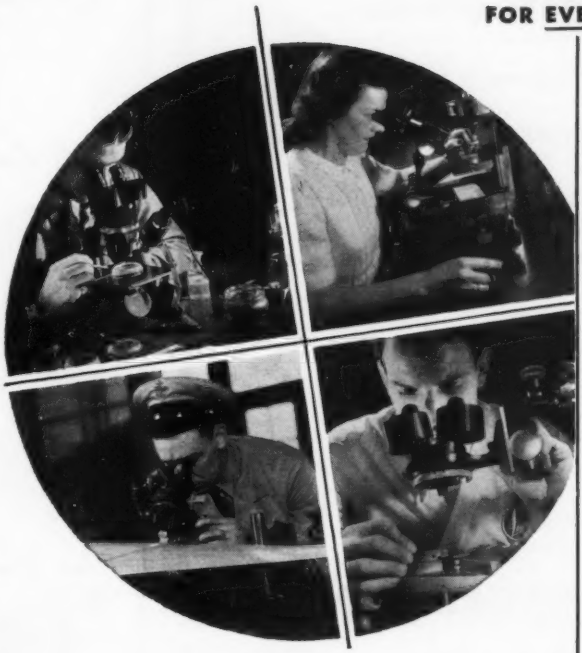

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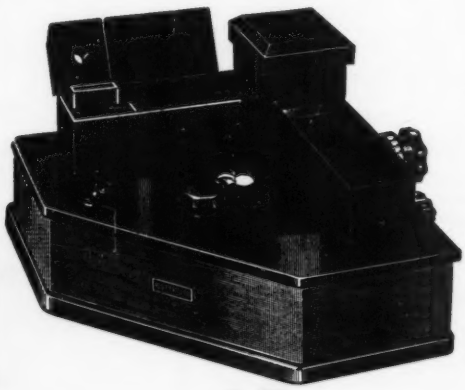

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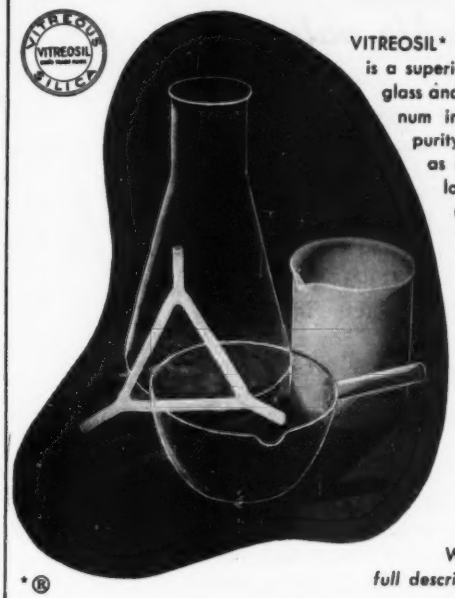

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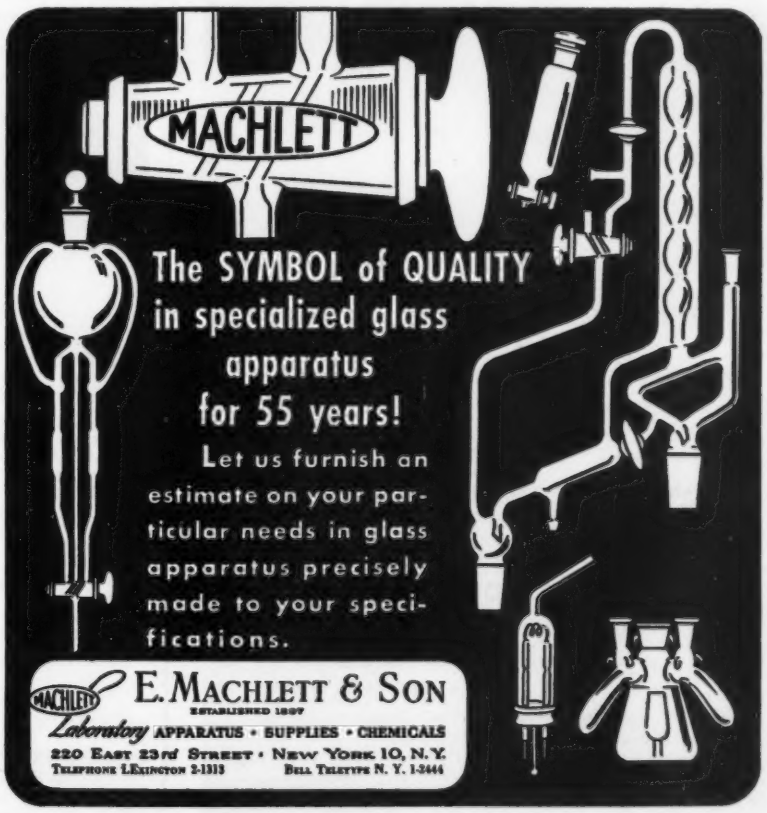

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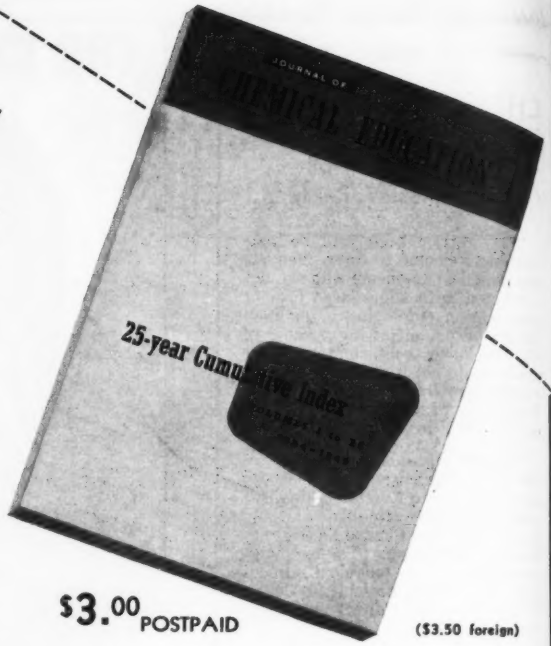

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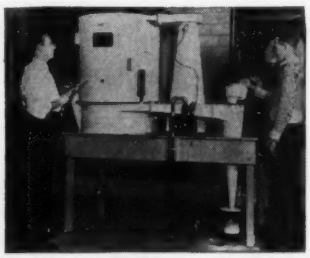

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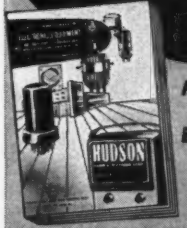

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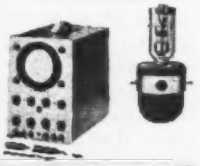

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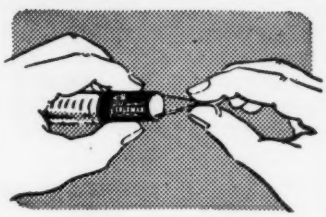
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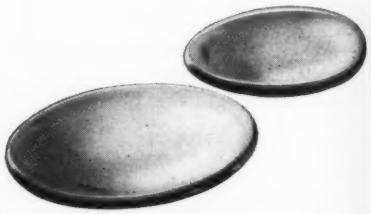

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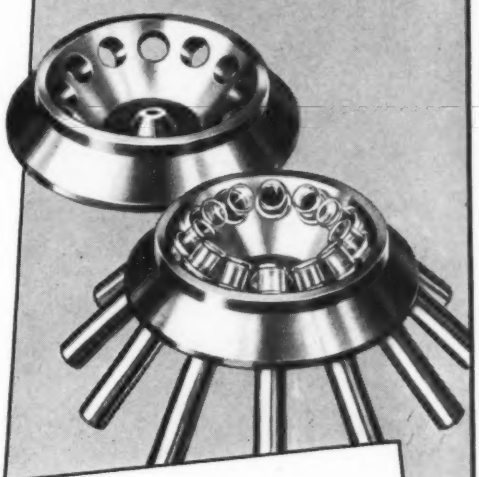

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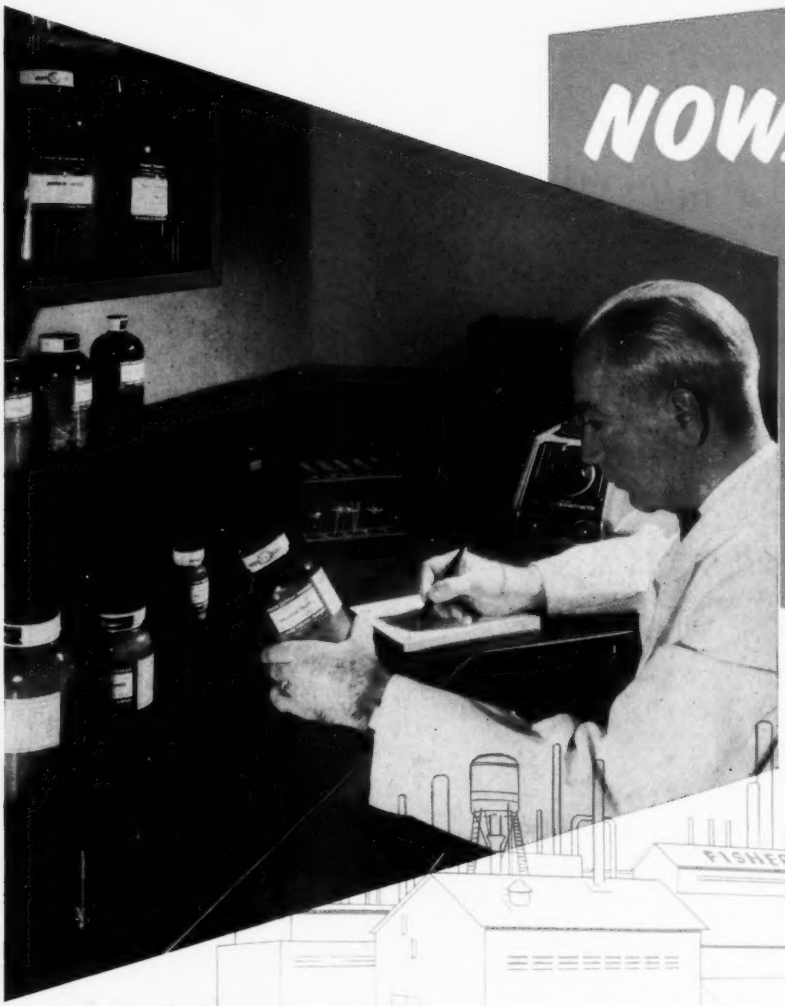

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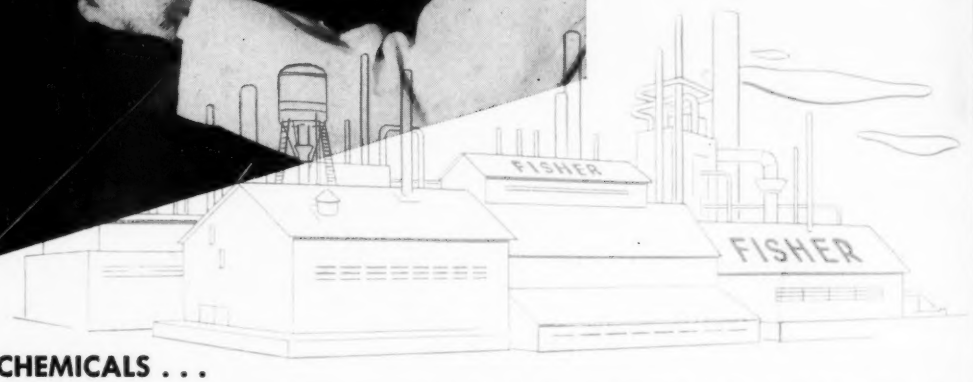

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